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CHEMICAL SEWAGE SLUDGE DISPOSAL ON LAND
(LYSIMETER STUDIES)
VOLUME II

by

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ABSTRACT

Lysimeter experiments in which crops were grown on agricultural soils treated with anaerobically digested sewage sludges (from plants where phosphorus removal was being practised using metal salts) were initiated in 1972 and 1973. Alum, iron and lime treated fluid sludges were added to loamy sand and silt loam soils vegetated with orchard grass. Similarly, alum, iron and lime treated air-dried sludges were added to sand and clay soils and wheat crops were grown. The high application rates of fluid and air-dried sludges ranged from 32 to 76 and 47 to 144 tons dry wt/ha/yr, respectively. The metal loadings approximated the recommended maxima for Ontario agricultural land. The lower rates were one-third and two-thirds of the high rates. The results of these experiments are presented and compared to results from field and greenhouse studies.

There was no evidence that the aluminum, iron or lime in the digested sludges was detrimental to crop production or plant tissue quality. Orchard grass yields from the fluid sludge experiment appeared to be related to nitrogen availability. The medium sludge application rates supplied about twice as much total Kjeldahl nitrogen (TKN) as the NPK (commercial fertilizer) treatment (control) but produced similar orchard grass yields. The residual effects of cumulative applications were evident as 22% of sludge treatment yields exceeded NPK in 1973 compared to 89% in 1976. No residual effect was observed where wheat crops were grown on sand. In 1974, wheat yields from 78% of sludge treatments exceeded NPK yields, compared to 0% in 1976.

The metal contents of crops grown on the sludged soils were generally less than, or equal to, suggested maximum tolerance levels. Zinc contents increased with increasing sludge application rates and tended to increase over the four-year experimental period at all sludge application rates. Copper contents increased as sludge application rates increased, but neither copper nor cadmium increased with time.

Trends in heavy metal uptake were similar for orchard grass grown in lysimeter experiments, brome grass grown in a field experiment, and rye grass grown in a greenhouse experiment. The sludges and

application rates were similar for the lysimeter and field experiments; however, the greenhouse experiments included treatments with high application rates of sludges containing high concentrations of various heavy metals.

Leachates from both experiments exhibited heavy metal concentrations well within drking water standards and showed no increase with time. Air-dried sludge applications resulted in nitrogen availability exceeding the crop requirements and leachates exceeding the 10 mg NO_3-N/l maximum permissible concentration (MPC) for drinking water. Phosphorus concentrations in leachates from the air-dried sludge experiment were always less than the 1 mg/l effluent MPC. This was also the case for the fluid sludge experiment through 1975. By 1976, leachate phosphorus concentrations from alum sludge applications exceeded the 1 mg P/l MPC.

In general, land application of sewage sludges at rates sufficient to supply the crop uptake of N and P would limit soil heavy metal loadings to acceptable levels for the sludges used in these lysimeter studies.

RÉSUMÉ

En 1972 et 1973, on a entrepris l'étude en lysimètre de la croissance sur des sols agricoles amendés avec des boues digérées en conditions anaérobies, provenant d'usines de traitement où l'on avait déphosphaté les eaux usées à l'aide de sels métalliques. On a ajouté des boues liquides traitées à l'alun, au fer et à la chaux à des sols gras sableux et limoneux cultivés en dactyle pelotonné. On a aussi ajouté des boues traitées de la même façon, mais séchées à l'air, à des sols sableux et argileux cultivés en blé. On a épandu les boues liquides et séchées à l'air à raison de 32 à 76 et de 47 à 144 t/ha/an (poids sec), respectivement. Leurs teneurs en métaux s'approchaient des quantités maximales recommandées pour les terres agricoles de l'Ontario. Les doses inférieures et médianes étaient respectivement égales au tiers et aux deux tiers des doses supérieures. Les résultats de ces expériences sont présentés et comparés à ceux d'études sur le terrain et en serres.

On n'a pu déceler aucun effet nuisible de l'aluminium, du fer ou de la chaux des boues digérées sur la production culturale ou la qualité des tissus végétaux. Le rendement du dactyle pelotonné semble relié à l'azote présent. Les doses médianes d'épandage fournissent environ deux fois plus d'azote total de Kjeldahl que le fertilisant commercial témoin NPK, mais donnent un rendement semblable en dactyle pelotonné. Les effets résiduels des épandages cumulatifs de boues nous apparaissent évidents; en effet, le rendement des boues dépassait celui du NPK dans 22 p. 100 des cas en 1973, comparativement à 89 p. 100 en 1976. On n'a noté aucun effet résiduel dans le cas des cultures de blé en sol sableux. En 1974, 78 p. 100 des amendements par boues ont permis une production de blé supérieure à celle des amendements avec le NPK, comparativement à aucun en 1976.

Les teneurs en métaux des cultures en sols traités avec des boues étaient généralement inférieures ou égales aux limites maximales de tolérance proposées. Les teneurs en zinc ont augmenté suivant

l'augmentation des doses d'épandage et ont eu tendance à s'accroître pendant les quatre années de l'expérience à toutes les doses utilisées. Les teneurs en cuivre on aussi augmenté suivant les doses d'épandage, mais ni la teneur en cuivre ni celle en cadmium ne se sont accrues avec le temps.

L'absorption des métaux lourds par le dactyle cultivé en lysimètre a suivi la même tendance que celle des cultures expérimentales de brome sur le terrain et d'ivraie en serres. Les types de boues et les doses épandues étaient semblables dans les expériences en lysimètre et sur le terrain. Les expériences en serres comprenaient toutefois des traitements à fortes doses de boues à concentration élevée de divers métaux lourds.

Les percolats engendrés dans les deux expériences contenaient beaucoup moins de métaux lourds que ne le prescrivent les normes pour les eaux potables, et on n'a décelé aucune augmentation avec le temps. Les boues séchées ont libéré plus d'azote que n'en requéraient les cultures, et des percolats dont la teneur en azote nitrique dépassait la concentration maximale permise (CMP) pour l'eau potable, soit 10 mg/l. La teneur en phosphore des percolats des boues séchés n'a jamais dépassé la CMP de 1 mg/l d'effluent. On avait obtenu le même résultat en 1975 avec des boues liquides. En 1976, cependant, la teneur en phosphore des percolats de boues traitées à l'alun dépassait la CMP de 1 mg/l.

En général, l'épandage sur le terrain des types de boues utilisés dans ces études en lysimètre, en quantité suffisantes pour fournir le N et de P nécessaires aux cultures, limiteraient les teneurs en métaux lourds des sols à des valeurs acceptables.

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SUMMARY AND CONCLUSIONS

Two separate lysimeter experiments conducted since 1973 were designed to determine long term effects on soils, plants, and leachate of applying:

I Fluid digested chemical sludges (alum, iron, and lime) to two medium textured soils growing orchard grass.

II Air-dried digested chemical sludges to clay and sand, growing wheat crops. The lysimeter experiment results are compared to data from:

- Ontario sludge disposal survey (1975).
- University of Guelph field and greenhouse experiments (1973 to 1975).
- Survey of plant metal uptakes from Ontario sludge farm sites (1976).
- Ontario Ministry of Agriculture and Food (OMAF) guidelines for sludge utilization on agricultural land.

1. Sludge constituent loadings (maximum rate) to lysimeter soils were 10 to 50 times the 1975 average annual loading to sludge farms in Ontario.
2. The OMAF sludge criterion relating the minimum acceptable ratio of soluble NH_4-N/heavy metals would allow use of fluid but not air-dried sludges. As nitrogen to metal ratios were higher, and metal concentrations lower in plant tissue receiving air-dried than fluid sludges, this criterion should be reconsidered for dry sludge applied to farmland.
3. At least 200% of NPK nitrogen rates must be applied as fluid sludge to obtain orchard grass yields equivalent to NPK treatment. The residual beneficial effect of cumulative sludge applications is evident as 89% of sludge treatment yields exceeded NPK in 1976 (loamy sand) compared to only 22% in 1973. Air-dried sludge wheat yields (sand) exceeded NPK yields in 78% of sludge treatments in 1974, but declined to 0%

in 1976. Wheat grain yields (1976) declined on sandy soil to 38% of NPK yields but increased to 103% of NPK from clay soil.

4. The heavy metals Cd, Cr, Cu, Ni, Pb, and Zn did not exceed suggested maximum "tolerance" or "toxic" concentrations in orchard grass or wheat crops (mean of all sludges 1973 to 1976). Metal concentrations in wheat grain were generally lower than in straw.

5. Only Zn was taken up by orchard grass or wheat crops in amounts exceeding the OMAF referenced maximum uptake of 0.3 kg/ha/yr. Zn and Cu uptake by orchard grass at the highest sludge rate was twice the NPK uptake rate. Ni, Cr, and Cd uptakes from sludges did not exceed NPK uptakes per equivalent dry matter yield. The uptake of metals in all crops receiving alum, iron or lime sludges reflect TKN uptake rather than sludge metal loading ratios.

6. Leachate volume losses from orchard grass soils varied inversely with crop yield and sludge application rate for each soil type. Leachate losses from air-dried sludge-wheat soils did not vary significantly with sludge loading rate.

7. Leachate maximum concentrations of Cd, Cr, Cu, Ni, Pb, and Zn did not exceed drinking water standards. Metals concentrations (mean of all sludges) were similar to NPK control. No increasing concentration trend for any metal monitored was observed during 1973 to 1976.

8. Nitrogen as NH_4-N rarely exceeded the drinking water MPC of 0.5 mg/l. The nitrate (NO_3-N) MPC of 10.0 mg/l was not exceeded during 1974 or 1975 (orchard grass) but increased during 1976 where alum and iron sludges (high rate) were applied to both soils. NO_3-N in leachate from lime sludge never exceeded 10.0 mg/l and was significantly lower than NO_3-N from alum or iron sludge applications. NO_3-N in leachate from air-dried sludge applications remained unacceptably high during 1974 to 1976. Control NO_3-N concentrations frequently exceeded 10.0 mg/l.

9. Soluble P in leachate exceeded 1.0 mg/l only from the high rate alum sludge treatment (loamy sand). Soluble P from all other sludge treatments to all soils generally met the drinking water MPC of 0.1 mg/l.

10. TOC in leachate from both experiments increased to >50 mg/l in 1976 at the highest sludge loading rates. Static bioassay toxicity tests using Daphnia showed no toxicity in leachate samples having high TOC values.

11. Limits related to water quality criteria are required for constituents of concern in leachate from sludge amended soils. Based on data from these lysimeter experiments, nitrate is the constituent of major concern in leachate, particularly when air-dried sludge is applied to a low nitrogen uptake crop. If leachate quality from normal agronomic (NPK) treatment is used as a standard of comparison, application of sludges at rates similar to NPK should not adversely affect groundwater quality. If drinking water criteria are adopted for leachate quality, commercial NPK application rates may also have to be regulated.

12. Recovery of NO_3-N in leachate as a percent of nitrogen applied to soil averaged 3% from fluid sludge and 33% from air-dried sludge at the highest rate. Heavy metal recoveries did not exceed 1% with the exception of cadmium and nickel. Nickel recovery from fluid sludge averaged 6.2% compared to 1.8% from air-dried sludge.

13. Brome grass crop yield trends from University of Guelph field experiments were similar to lysimeter orchard grass yield response to sludge treatments.

14. Plant tissue heavy metal concentration trends (highest rates of alum, iron, and lime sludges) were generally similar for both lysimeter and field experiments. Cadmium did not increase in either field, greenhouse or lysimeter experiments using similar sludge sources. Copper increased somewhat in both lysimeter and field experiments, but did not exceed the suggested toxic concentration (20.0 μg/g). Rye grass grown in the greenhouse showed some yield depression attributed to Cu

toxicity. Nickel did not exceed the suggested tolerance concentration of 3.0 µg/g in either lysimeter or field experiments. Even concentrations of Ni in excess of 40 µg/g in rye grass (greenhouse) did not show any apparent yield depression effect. Zinc increased slightly in field experiments but not in lysimeter experiments. Rye grass Zn concentrations above the suggested toxic level of 200 µg/g did not show any yield depression effect. Lead concentrations did not increase in either lysimeter, field or greenhouse experiments. High Pb values in lysimeter experiment crops are attributed to aerial contamination.

15. Heavy metal concentration trends in plant tissue from the lysimeter experiments were compared with metal concentrations in crops from selected sludge farms and adjacent non-sludge control sites in Ontario. The sludge farms were selected on the basis of high sludge metals loadings over extended periods of time. The constituents analyzed included As, Cd, Co, Cr, Cu, Hg, Mo, Ni, Pb, Zn, and PCB's. Although many of the heavy metals exceeded the OMAF maximum recommended content in soils, concentrations in plant tissue from sludged sites were generally not significantly greater than concentrations of these metals in plant tissue from control sites. The only exception was Mo, with one sludged site exhibiting a potentially toxic concentration of Molybdenum in corn stover (24 µg/g).

16. More information is required concerning cumulative uptake of metals from a wide variety of agricultural crops, receiving sludge in either fluid or dry form. More information concerning the tendency of a particular crop to remove a particular "constituent of concern" from sludge amended soil with a high recovery efficiency could allow for increasing refinement in sludge application guidelines as currently proposed.

17. If nitrate pollution of groundwater as a result of sludge application to soil becomes a major factor limiting sludge application rates, similar restrictions in application of

commercial NPK fertilizers might be anticipated. Background information concerning the seasonal variability of NO_3-N concentrations in groundwater where no fertilizer is applied at all would be of benefit in evaluating the impact of sludge loadings on groundwater quality.

18. In view of the non-lethal toxicity results obtained to date from leachate samples having high TOC values, carbon chloroform extract (CCE) tests will, in future, be compared to TOC results to provide a more sensitive indication of potentially deleterious organic substances.

19. Fluid lime sludge application even at the highest rate (unlike alum and iron sludges) did not result in unacceptable NO_3-N leachate levels in either soil during four years of monitoring. Futher research is required to determine whether this differential nitrate formation is due to inhibition of soil nitrifying bacteria or accelerated gaseous nitrogen losses as NH_3 or N_2.

1 INTRODUCTION

A survey of sludge disposal practices in Ontario by Antonic et al (1978) indicates substantial increases in both sludge volumes (14 to 70%) and dry weight (34 to 113%) as a result of chemical additions to wastewater for phosphorus removal. The chemicals most commonly used for this purpose are aluminum sulphate (alum), ferric chloride or waste pickle liquor (iron), and calcium oxide or hydroxide (lime). Little information is available concerning the environmental impact of these sludges in either liquid (fluid) or dry forms when applied to a wide variety of agricultural soils producing many different crops. Of particular concern to public health authorities and regulatory agencies are the potentially deleterious effects on soils, groundwater, and plants of the numerous toxic substances such as heavy metals which are increasingly concentrated in wastewater sludges as effluent regulations become more stringent.

Ultimate disposal of sludges requires evaluation of the relative benefits and risks of utilizing the three major disposal sinks available; namely the atmosphere, the oceans or the land. A paper by Ember (1975) indicates that the risks of ocean disposal outweigh the potential benefits. Aquatic organisms tend to accumulate metals and other toxic constituents in extremely high concentrations. For example, a cadmium biomultiplication factor of 14,400 has been reported for oysters. Land disposal of sludges, therefore, appears to be a safer alternative than ocean disposal because soils have the capacity to tie up trace elements thus making them less available to land plants than they are to aquatic organisms.

Incineration, another alternative to land disposal, is the major ultimate disposal method used in Ontario, accounting for 40% (62,000 tons per year) of all sludge disposed of in the province. The atmosphere is not intentionally used as a sink for heavy metals, when disposing of sludge in this manner. A report by Van Loon (1976) indicates that at the incineration temperatures generally used for sludge disposal (700 to 900°C) substantial amounts of cadmium, zinc, arsenic, lead and mercury will be volatilized, thus potentially recontaminating soil, water, plant, and aquatic life in the "fallout" zone.

Application of sewage sludge to farmland in Ontario (1975) accounted for 34% (53,000 tons/year) of all sludge disposal in the province. The benefits to the treatment plants involved are relative simplicity and low cost compared to the other alternatives. The farmers benefit in the short term from a free source of essential plant nutrients and organic matter, but the long term risks of toxic constituent accumulations in soil, plants, and groundwater remain to be determined.

Land application of sludges is frequently the lowest cost ultimate disposal alternative. Black and Schmidtke (1974) indicated that fluid sludge haulage costs ranged from \$21 to \$275 per ton dry solids in Ontario in 1973. In 1975, the average cost for hauling fluid sludge in Ontario was \$1.63 yd^3. The high costs of hauling fluid sludge provide an economic incentive for determining the maximum acceptable sludge loading rates to land. They also provide an incentive for investigating more economic methods of sludge dewatering or drying to avoid paying for transporting the 90 to 99% water contained in the fluid sludge.

The long term risks of applying chemical sludges to farmland must be assessed. These estimates of risk should then be incorporated into guidelines or standards which determine with reasonable factors of safety a permissible code of practice. The standards should state clearly the maximum permissible concentrations of toxic or undesireable contaminants in sludge, soil, plants, runoff, and leachate to groundwater. These "ideal" standards must be based on an acceptable baseline of scientific information, much of which is unfortunately sorely lacking. The attempt to fill in some of the above mentioned information gaps by carefully controlled long term lysimeter studies monitoring soil, sludge, plant, and leachate quality to obtain material balances for the various "constituents of concern" is the subject of this report.

2 OBJECTIVES

The major objective of these studies is to determine the maximum sludge application rates which can be applied to various agricultural soils growing either forage or edible crops (wheat) without causing deleterious effects to:

- soil productivity (crop yield);
- plant quality (heavy metal toxicity); and
- groundwater quality (for drinking or irrigation).

Secondary objectives include a comparison of the lysimeter results with:

- field and greenhouse studies - University of Guelph;
- Ontario provisional guidelines for sludge utilization on agricultural lands;
- survey of Ontario sludge disposal practices; and
- survey of heavy metals uptakes from selected farm sites in Ontario to which sludge has been applied.

3 EXPERIMENTAL DESIGN

Two separate lysimeter experiments were initiated at the Wastewater Technology Centre, Burlington, one year apart. The first, begun in 1972, was designed as a randomized block of 66 lysimeters with two soil types, three replications, and eleven fertility treatments consisting of three sludge types, three sludge rates and two controls (with and without NPK commercial fertilizer). The sludge consistency (fluid), soil types (medium texture), and agricultural crop (orchard grass), were chosen to permit subsequent comparison with the most common practice of sludge disposal to agricultural land in Ontario. A perennial grass crop was used for maximum potential nitrogen uptake. Sludge loading rates (kg TKN/ha) were chosen to closely approximate the crop soluble nitrogen requirement at the lowest rate, with the medium and high rates double and triple the low rate. Sludge types were chosen to include the three most common chemicals used for phosphorus removal (alum, iron and lime).

The second experiment, begun in 1973, was designed as a randomized block of 44 lysimeters, eleven fertility treatments replicated twice on two different soils. Air-dried (∿5% moisture) sludge rather than fluid sludge was used to assess the provisional OMAF guidelines (1976) which are based on a soluble NH_4-N to metals ratio. Air-dried sludges containing less than 1% of TKN in soluble form are, theoretically, unsuited for application to farmland. The high cost of fluid sludge haulage was another reason that dry sludge was chosen. The crop selected was wheat (spring and fall) to permit a comparison of metals uptake in both the grain and leafy portions of the plant. Sludge loadings were based on Chumbley's (1971) zinc equivalent recommendation of no more than 250 mg/kg zinc equivalent metal addition to soil to prevent toxicity to plants. Actual sludge loadings at the end of the first year were 6, 12, and 18 times the maximum zinc equivalent loading recommended by Chumbley.

Two widely divergent soils were chosen for this experiment, namely a sand soil having zero clay and only 3.0% silt in the "A" horizon, and a clay type soil with 33% clay and 59% silt. Sludge was applied in 1974, but not in 1975 or during the 1976 growing season.

4 MATERIALS AND METHODS

4.1 Lysimeters

The first experiment (fluid sludge-orchard grass) used 30 cm diameter, 1.8 m deep cylindrical fibreglass lysimeters, housed in an environmentally controlled trailer. The tops of the lysimeters projected 7.6 cm above the roof of the trailer and were subjected to normal ambient and atmospheric conditions. A temperature control unit was installed inside the trailer to simulate field winter temperatures (4±1°C) and ambient soil temperatures during the summer. Facilities were provided at the bottom of the lysimeters for leachate collection by gravity or by vacuum extraction at several depths within the soil profile.

The second experiment (air-dried sludge-wheat) used square polyethylene lysimeters of larger surface area (60 x 60 cm) and less depth (76 cm) for the annual cereal crop. These lysimeters were exposed to ambient conditions throughout the year.

4.2 Soils

The two soils chosen for the first experiment were identical to soils used by the University of Guelph for their field trials. The textural class names based on particle size classification are:

Loamy sand (from Hespeler, Ontario).
Silt loam (from Elora, Ontario).

A brief description of these two soil types is given in Table 1.

At both soil acquisition sites, horizons A, B, and C were collected separately. Soils were rebuilt in the lysimeters as close as possible to the original field density by adding the soil in layers of about 7 cm. All lysimeters were saturated with water from the bottom to remove air pockets and then gravity drained. The lysimeters were then placed in the trailer for about six months to allow complete soil settling before crop establishment commenced in May, 1973.

TABLE 1. SOIL TYPES USED IN FLUID SLUDGE-ORCHARD GRASS EXPERIMENT

Soil Type	Horizon Depth (cm)	pH	Organic Matter %	Sand %	Silt %	Clay %	Bulk Density g/cm^3
Loamy Sand	A-19	7.8	2.4	74	23	3	1.4
	B-15	7.5	0.9	73	27	0	1.6
	C-87	8.6	0.6	92	8	0	1.6
Silt Loam	A-19	7.8	3.6	43	44	13	1.2
	B-15	8.0	1.6	42	42	16	1.5
	C-45	8.6	0.6	40	49	11	1.7

The two soils chosen for the second experiment were as divergent as possible from the soils of the first experiment to provide a broad range of soil types for comparison. Figure 1 shows the textural range of all four soils used in both experiments, based on particle size classification. The textural class names of the soils used in the air-dried sludge-wheat experiment are:

Clay (from New Liskeard, Ontario).
Sand (from Norfolk County, Ontario).

Both soils were used for growing pasture grass prior to acquisition for this experiment. The sandy soil is developed from excessively drained dune sand, and is subject to wind and water erosion. The clay soil is developed on lacustrine deposits and is poorly drained.

A brief description of these two soil types is given in Table 2.

The two soils had similar horizonal depths. Horizons A, B, and C were collected separately, as in the first experiment, and the soils were then rebuilt in the lysimeters by packing in 5 cm layers to their original field bulk density. Water was supplied from the base drain to remove air pockets, and the soils were allowed to stabilize under ambient conditions for about four months, before crop seeding in May, 1974.

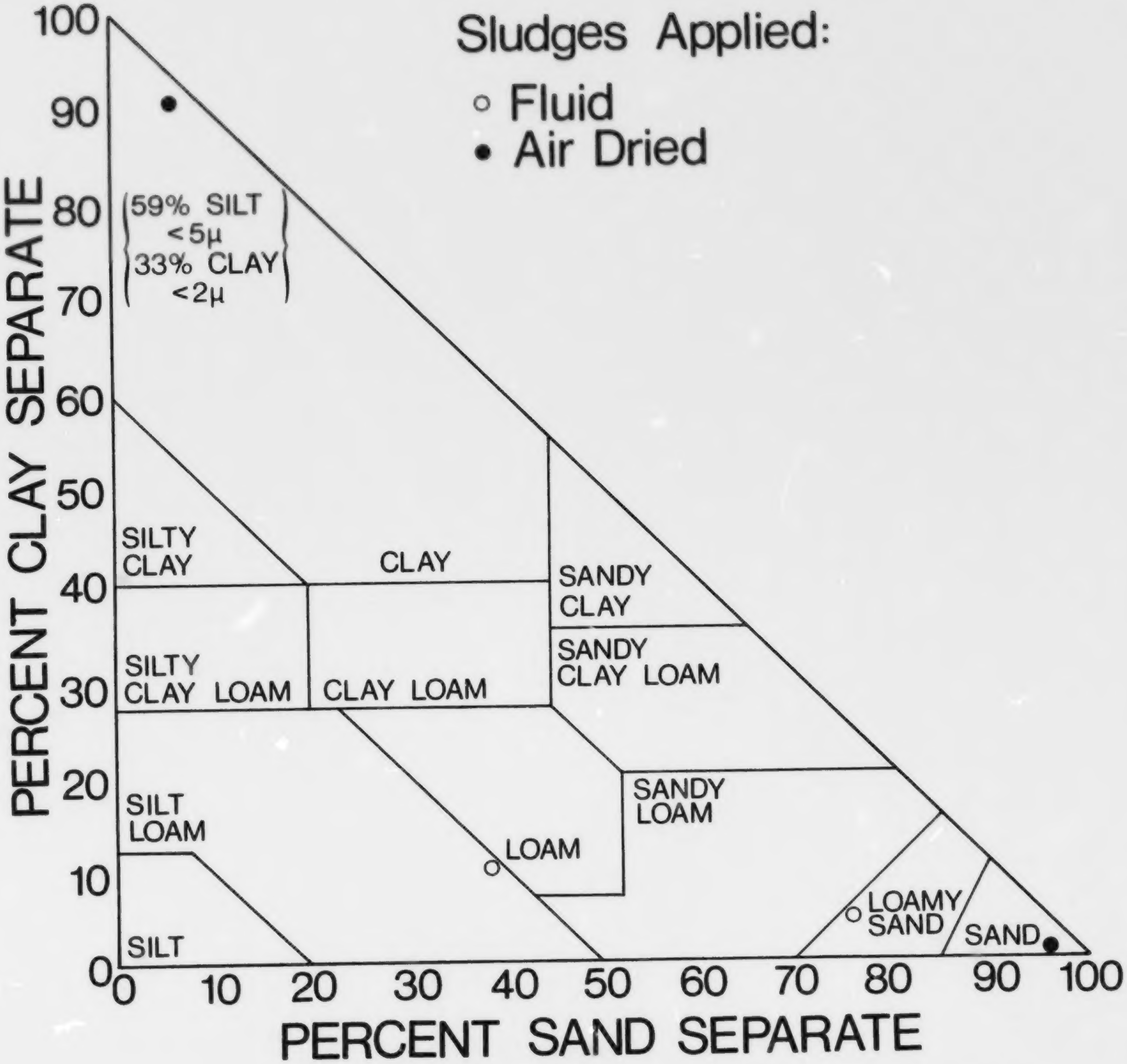


Figure 1 SOIL TEXTURAL CLASSES BASED ON THE PARTICLE SIZE CLASSIFICATION

TABLE 2. SOIL TYPES USED IN AIR-DRIED SLUDGE-WHEAT EXPERIMENT

Soil Type	Horizon Depth (cm)	pH	Organic Matter %	% Particle Size			Bulk Density g/cm^3
				Sand %	Silt %	Clay %	
Sand	A-15	5.6	2.1	97	3	0	1.5
	B-10	6.3	1.4	91	9	0	1.6
	C-25	6.7	0.5	98	2	0	1.6
Clay	A-15	7.2	6.4	8	59	33	1.2
	B-10	7.3	2.1	3	45	52	1.5
	C-25	7.5	0.9	4	23	73	1.7

4.2.1 Soil chemical analysis

The tentative guidelines for sludge disposal to agricultural land in Ontario have established criteria for maximum allowable concentrations of total heavy metals in sludge amended soils, based on the mean metal content in uncontaminated soils (Frank et al, 1976). To compare the "before" and "after" effects of sludge addition to the four soils used in the lysimeter experiments, 15 chemical constituents were analyzed in each separate soil horizon according to standard methods (Black, 1965). The results of some of these analyses (A horizon soils only) are compared with the OMAF guideline mean metal contents in Table 3.

4.3 Sludges

Attempts were made to correlate the sources of alum, iron and lime anaerobically digested sludges, used in the lysimeter experiments, with those used in field experiments at the University of Guelph. Changes in the chemical used for phosphorus removal at the water pollution control plants (WPCP), coupled with timetable and transportation logistic problems made this objective unattainable. The ferric chloride treated sludge from North Toronto, and the lime treated sludges from either Newmarket or Midland compare reasonably well in chemical composition with the iron and lime sludges used by the University of Guelph. The alum sludges were obtained from Point Edward, Tillsonburg, and Kitchener

TABLE 3. SOIL CHEMICAL ANALYSIS (mg/kg dry weight) OF "A" HORIZON SOILS USED IN LYSIMETER EXPERIMENTS - COMPARED TO OMAF GUIDELINES MEAN METAL CONTENT IN UNCONTAMINATED AND SLUDGE AMENDED ONTARIO SOILS

Constituent	Sand	Clay	Loamy Sand	Silt Loam	Metal Content Uncontaminated[1] Ontario Soil	
					Mean	Max.
TKN	1,500	3,800	1,150	2,000	-	-
Total P	700	900	1,520	2,660	-	-
Soluble P	NA	NA	0.8	0.5	-	-
Total Ca	15,000	14,500	17,000	19,000	-	-
Total K	10,000	22,100	15,000	17,000	-	-
Total Fe	9,200	29,500	23,000	34,000	-	-
Total Al	22,800	80,000	45,000	48,000	-	-
Total Zn	26	100	105	100	54	216
Total Cu	4	26	13	20	25	100
Total Ni	6	40	9	17	16	32
Total Cr	15	63	27	45	14	56
Total Pb	10	23	18	16	14	56
Total Cd	0.7	1.0	0.1	0.1	0.7	1.4
CEC meq/100 g	4.7	28.2	9.0	18.2	-	-

[1] Source: OMAF Provisional Guidelines for Sewage Sludge Utilization on Agricultural Lands - June, 1976.

NA = not available.

and differed markedly among themselves. Consequently, sludge constituent loadings to soils from alum sludges, particularly for some heavy metals, are different in the lysimeter experiments than those in the University of Guelph field studies.

The fluid sludge sources used in the first experiment were also the original sources of the air-dried sludges used in the second experiment. The desired moisture content (<5% H_2O) was obtained by mechanically dewatering the fluid sludge and spreading the sludge cake on plastic sheets to dry at ambient temperature.

The chemical composition of typical sludge sources collected in 1974 are presented in Table 4.

TABLE 4. SLUDGE CHEMICAL COMPOSITION (1974 - TYPICAL) DRY WEIGHT BASIS

Constituent	Fluid			Air-Dried		
	Alum	Iron	Lime	Alum	Iron	Lime
TKN %	4.9	5.0	4.1	3.7	3.1	1.7
NH_4-N %	1.2	1.3	1.0	0.09	0.05	0.01
Total P %	4.6	3.4	2.6	4.9	2.0	0.9
Total Ca %	23.5	7.8	44.2	5.7	5.0	23.0
Total Fe %	1.8	8.2	2.4	1.0	6.0	0.6
Total Al %	18.1	2.4	4.2	6.6	1.1	0.3
K mg/kg	1474	1424	1515	750	600	417
Zn "	868	2397	1242	1050	1800	290
Cu "	816	1534	515	539	858	140
Ni "	11	14	12	10	15	8
Cr "	168	562	52	165	510	37
Pb "	263	1740	303	280	1480	119
Cd "	13	26	15	12	22	5

4.3.1 Sludge loading rates

The two major criteria in the OMAF sludge guidelines determining the maximum rate of sludge which can be applied, in any one five-year period, to a particular soil are:

- nitrogen requirement of the crop; and
- initial metal content in soil.

The first experiment was designed to assess the nitrogen criterion. Cumulative multiples of the baseline recommendation, i.e., 120 lb/acre (134.5 kg/ha) of soluble NH_4-N were applied annually. The yield of orchard grass which could be harvested during a particular growing season was determined by the cumulative application rate based on total Kjeldahl nitrogen (TKN). Fluid sludge was applied three times in 1973, four times in 1974, and five times in 1975, and 1976, each time at rates of 100, 200, and 300 kg TKN/ha. By the end of 1975, 3,600 kg TKN/ha had been applied at the highest rate. The soluble NH_4-N loading ranged from 800 to 1,000 kg N/ha (22 to 29% of TKN).

The second experiment was designed to assess the heavy metals criterion. Using the Chumbley (1971) "zinc equivalent" formula based on the relative toxicity to plants of zinc, copper and nickel $[\Sigma \text{ Zn} + 2 (\text{Cu}) + 8 (\text{Ni})]$ over a 30-year period, the maximum recommended zinc equivalent (ZE) was 19 kg (ZE)/ha/year. The air-dried sludge was applied on two occasions in 1974 to the surface 15 cm (A horizon) of each lysimeter at cumulative rates of 114, 228, and 342 kg (ZE)/ha. The maximum loading rate is 18 times the maximum annual application rate recommended by Chumbley. In order to determine the residual effect of a single annual loading of heavy metals to the two soil systems under investigation, no sludge was applied during either the 1975 or 1976 growing season.

Table 5 shows the cumulative loading rates (kg/ha) of the various chemical constituents for both the fluid and air-dried sludge experiments, up to the end of the 1975 growing season at the highest application rate.

4.4 Crops

The need for a forage crop with a high nitrogen removal efficiency (>200 kg N/ha/yr), minimal genetic variability between seedlings, hardiness, and adaptability to most agricultural soils led to the choice of orchard grass, Frode variety (Dactylis glomerata L.), for the fluid sludge lysimeter experiment. After harvesting, grass samples were washed with tap water and rinsed with distilled water to remove surface adsorbed contaminants, oven dried at 70°C, ground to pass through a 60 mesh screen and chemically analyzed for nutrients and heavy metals, using standard plant analysis procedures (Walsh, 1973).

TABLE 5. SLUDGE CONSTITUENT LOADINGS AT THE HIGHEST LOADING RATE* (kg/ha)

Constituent	Fluid Sludge (1973+74+75)			Air-Dried Sludge (1974 only)		
	Alum	Iron	Lime	Alum	Iron	Lime
TKN-N	3600	3600	3600	3558	2487	6984
NH_4-N	906	1035	807	69	21	36
Total P	3363	1899	1842	5391	3072	8052
Ca	8577	4608	44859	7488	7038	114102
Fe	900	6357	1920	1242	5805	3087
Al	8724	1437	1422	7557	840	1101
K	117	174	156	114	60	165
Zn	91	212	155	154	163	174
Cu	59	102	45	80	84	71
Ni	2.4	2.1	3.0	3.4	1.2	3.3
Cr	15	46	7	19	41	14
Pb	25	135	29	36	117	47
Cd	0.8	2.1	1.3	1.5	2.3	1.5
Total Solids (tons/ha)	91.2	82.2	175.5	140.1	95.4	430.5

* Lowest loading = 1/3; medium loading = 2/3 the values listed.

The crops chosen for the air-dried sludge experiment included: spring wheat (Triticum aestivum) Glenlea variety; fall wheat (Triticum aestivum) Genesee variety; and buckwheat (Fadopyrum esculetum) as a summer cover crop between wheat crops to prevent weed spread and to minimize nitrogen loss in the leachate. The spring wheat seeded in May, 1974 was harvested in August, 1974. The plants were clipped 2 cm from each end of the lysimeter to minimize border effects. The net harvested area was 0.35 m^2/lysimeter. After harvesting, the crop was threshed, with grain and straw portions dried, ground, screened and analyzed separately using methods of the Analytical Services Section, Wastewater Technology Centre, Burlington (Conn, 1976). Fall wheat was seeded in September, 1974 and harvested in July, 1975. Buckwheat was seeded in July, 1975 and harvested in September, 1975. A second cycle of spring and fall wheat was begun in 1976.

4.5 Leachate

In addition to normal precipitation, the lysimeters were irrigated as required to maintain soil moisture at one-third field capacity or greater. Equal volumes of water were supplied to all treatments in each experiment. Leachate collection periods were variable in duration and frequency depending upon changing climatic conditions. Volumes collected during both the winter frozen soil period and the summer growing season (when evapotranspiration losses were high) were less than during the spring "thaw" period, and at the end of the summer growing season.

For the fluid sludge-orchard grass experiment, leachate collection coincided with grass harvest and preceeded sludge application dates. Field measurement of volume, pH and conductivity preceeded filtration through a 0.45 micron filter. The samples were stored at 4°C until analyzed according to Standard Methods (APHA, 1971).

For the single application air-dried sludge-wheat experiment, leachate collection periods were based on plant development. Period 1 coincided with crop emergence, Period 2 was from emergence to the fifth leaf stage, Period 3 was from fifth leaf until harvest, and Period 4 from harvest until emergence of the next crop.

5 RESULTS AND DISCUSSION

5.1 Soils

Many factors that influence availability and uptake of trace metals and other constituents of concern from soils, including clay and humic content, both of which affect the cation exchange capacity (CEC) of the soil are mentioned in the literature.

Although the levels and specific chemical forms at which toxicity in soils occurs will vary from soil to soil and have not yet been established, Ontario provisional guidelines do place limits on specific heavy metals in soils to guard against buildup to levels which have been implicated as causing phytotoxicity in Europe (Rhode, 1962).

The four soil types used in these lysimeter experiments do not conform to the OMAF provisional guideline recommendations in the following respects:

1. Soil pH - the value of 6.0 below which sludge is prohibited from being applied to agricultural soil was not met in "A" horizon of the Norfolk sandy soil (pH 5.6). This soil should, therefore, be particularly scrutinized for increased metal availability and solubility in plants and/or leachate compared with the three other neutral to alkaline soils.
2. The concentrations of nickel and chromium in the New Liskeard clay soil exceeded the maximum recommended metal content in soil, of 32 and 56 mg/kg, respectively. The concentration of cadmium (1.0 mg/kg) is almost equal to the maximum recommended content (1.4 mg/kg). Counterbalancing the high metal concentrations are the high organic matter contents (6.4%) and cation exchange capacity (28.2 meq/100 g) which tend to tie up metals and make them unavailable for plant uptake. These metals (Ni, Cr, Cd), should be particularly scrutinized for increased concentrations with time in plants and/or leachate. Soil scientists in the U.S., such as Allaway (1968) report common ranges of trace elements typically

found in unsludged soils which greatly exceed the maximum metal content recommended in the OMAF provisional guidelines for Ontario.

5.2 Sludges

A report on sludge disposal practices in Ontario by Antonic et al (1978) summarizes the mean content of heavy metals in sludges from 40 WPC plants in Ontario.

To determine the potential contribution of industry to the heavy metal problem these values, and the range of values for lysimeter experiment sludges, are compared with the metal content of sludges from the Chicago Calumet WPCP before and after passage of an industrial wastewater discharge ordinance in 1969 (McCalla et al, 1977) in Table 6.

The range of values observed for trace metals in sludges applied to the lysimeter experiments are all "normal" by Ontario standards, with the exception of nickel and chromium. The Calumet values are all significantly higher before passage of the 1969 ordinance but, by 1974, the trace element concentrations dropped below the Ontario range for metals such as chromium, copper, nickel and lead. The possibility of reducing the very high values of heavy metals in some Ontario municipal sludges (Antonic et al, 1978) to levels which would be acceptable for application to farmland, by means similar to those pursued by the Metropolitan Sanitary District of Chicago, is intriguing.

Based on the mean annual loading rate in Ontario of 4.9 tons/ha (dry weight), Antonic et al (1978) have calculated the amounts of nutrients and heavy metals that a sludge of "average" composition would contribute to uncontaminated soil. Table 7 compares these values from the Ontario sludge disposal survey with the highest loadings applied to the lysimeters from either fluid or air-dried sludges. Virtually all nutrients and metals (with the exception of K and Ni) were applied to the lysimeter experiment at rates 10 to 50 times greater than "average loading" in Ontario. Because cumulative applications are still being applied to these lysimeters (particuarly fluid sludge to orchard grass), confidence in the applicability of this information concerning safety factors

TABLE 6. COMPARISON OF TRACE ELEMENT CONCENTRATIONS OF WTC SLUDGES WITH ONTARIO AVERAGE VALUES (ANTONIC et al, 1978) AND CALUMET-CHICAGO VALUES (McCALLA et al, 1977) (mg/kg dry weight)

Constituent	Ontario Secondary Plants (Average) *	WTC - Lysimeter Study (Range)	Calumet - Chicago (Average)	
			Before Ordinance	After Ordinance
			(1969)	(1974)
Cd	30	6 - 26	190	54
Cr	1200	40 - 562	2100	790
Cu	810	140 - 1554	1500	282
Ni	170	8 - 15	1000	77
Pb	600	130 - 1740	1800	486
Zn	1480	290 - 2400	5500	2800

* From 40 WPCP's.

TABLE 7. COMPARISON OF SLUDGE CONSTITUENT AVERAGE LOADINGS TO ONTARIO FARMLAND (Antonic et al, 1978) WITH MAXIMUM LOADINGS TO WTC LYSIMETERS (kg/ha)

Constituent	Ontario Survey Annual Average (1975)	WTC Lysimeter Experiments Maximum Cumulative Loadings		Ratio $\frac{\text{WTC Loading}}{\text{Ontario Average}}$	
		Fluid Sludge (1973-75)	Air-Dried Sludge (1974)	Fluid	Air-Dried
N	252	3600	6984	14.3	27.7
P	126	1121	8052	8.9	63.9
K	24	174	165	7.3	6.9
Zn	6.6	212	174	32.1	26.4
Cu	4.1	102	84	24.9	20.5
Ni	0.8	3.0	3.3	3.8	4.1
Cr	4.9	46	41	9.4	8.4
Pb	2.6	135	117	51.9	45.0
Cd	0.17	2.1	2.3	12.4	13.5

TABLE 8. NUMBER OF YEARS REQUIRED TO REACH OMAF MAXIMUM RECOMMENDED METAL CONTENT IN LYSIMETER SOILS AT THE HIGHEST SLUDGE LOADING RATE

Constituent	Loamy Sand (Fluid Sludge)		Clay (Air-Dried Sludge)	
	Initial Content (mg/kg)	Time to Reach Maximum Recommended Content (years)	Initial Content (mg/kg)	Time to Reach Maximum Recommended Content (years)
Cu	13	6.7	26	3.6
Zn	105	4.3	100	2.6
Ni	9	76.7	40	<1.0
Cr	27	5.4	63	<1.0
Pb	18	2.1	23	1.1
Cd	0.1	9.3	1.0	1.3

TABLE 9. COMPARISON OF OMAF GUIDELINE MAXIMUM ACCEPTABLE METAL CONCENTRATIONS IN DRIED/COMPOSTED SLUDGES (<5% NITROGEN) WITH MAXIMUM CONCENTRATIONS IN AIR-DRIED SLUDGE APPLIED TO WTC LYSIMETERS (mg/kg dry weight)

Constituent	OMAF Maximum Recommended Concentration	Air-Dried Sludge Maximum Concentrations	Ratio $\frac{\text{WTC Sludge Maximum}}{\text{OMAF Recommended Maximum}}$
Cd	3.5	22	6.3
Cr	210	510	2.4
Cu	375	858	2.3
Ni	80	15	0.2
Pb	210	1480	7.0
Zn	810	1800	2.2

required to avoid deleterious environmental effects to these soil-crop systems should increase with each passing year.

The OMAF provisional guidelines include a recommended maximum metal content in the plough zone layer of sludge amended soils. Table 8 summarizes the number of years required to reach the OMAF maximum metal content for the fluid sludge applied to loamy sand and the air-dried sludge applied to clay at the highest sludge loading rate. As the fluid sludges have been applied annually to the same lysimeter soils since 1973, the maximum recommended metal content will be reached during 1977 for Zn, Cr and Pb, while the maximum recommended content for Cu will be reached in 1978/79.

The high concentration of trace metals in the clay soil following 1974 sludge applications resulted in nickel, chromium, lead, and cadmium exceeding the maximum recommended content, in 1975. Application of additional sludge at the end of the 1976 growing season has increased the metal content of the clay soil above the maximum recommended concentration for most of the heavy metals being analyzed.

The only reference in the OMAF (1976) provisional guidelines to dried sludge maximum acceptable metal concentrations appears in Appendix 1 titled "Dried and Composted Sewage Sludges" for home gardens, lawn or golf course applications. Table 9 compares these maximum recommended concentrations with the actual maximum sludge metal concentrations in the air-dried sludges used in the lysimeter experiment. With the exception of nickel, all metals analyzed had from two to seven times greater metals concentrations than those recommended in this guideline.

The Ontario provisional guidelines do not consider differences in soil properties when recommending maximum metal additions to soil. The U.S. land grant universities and the USDA (Ryan, 1976) have, however, considered these differences in recommending to U.S. Environmental Protection Agency (EPA) interim limits on metals application to agricultural soils. Table 10 uses soil cation exchange capacity as a method of grading soils for their affinity to metals until more information becomes available concerning the chemistry of metals in sludged soils. The USDA guidelines were compared to sludge loadings to the four soils used in the WTC lysimeter experiments. The amounts of zinc and copper applied to the sandy soil (CEC = 4.7 meq/100 g) between 1973 and 1976 exceed the maximum cumulative amounts recommended, while cadmium equals 70% of the maximum recommended application. Conversely, if the 1974 loading were repeated annually, it would take 24 years to reach the maximum recommended loadings of zinc and copper to the clay soil. The wide range in lysimeter sludge loading conditions (based on either Ontario or U.S. recommended guidelines) should help to provide, within a reasonable period of time, information required to develop a broader data base for regulating sludge application to farmland. Periodic review of this information may determine whether safety factors built into existing guidelines should be maintained or altered.

TABLE 10. MAXIMUM CUMULATIVE SLUDGE METAL APPLICATIONS (kg/ha) FOR PRIVATELY OWNED LAND (USDA RECOMMENDATION)[1]

Metal	Soil Cation Exchange Capacity (meq/100 g)[2]		
	0 - 5	5 - 15	>15
Zn	250	500	1000
Cu	125	250	500
Ni	50	100	200
Cd	5	10	20
Pb	500	1000	2000

[1] Federal Register, Vol. 41 - No. 108, June 3, 1976, page 22533-22536.
[2] Determined on unsludged soil.

5.3 Crops

A literature review by Page (1974) indicates that sewage sludges from most domestic WPC plants can be applied in modest amounts to most agricultural soils for limited periods without adversely affecting yields or quality of most crop species. The dangers of plant or food chain toxicity due to trace element soil enrichment may increase, however, if sludges are applied:

- to inappropriate soils (e.g., low pH);
- from highly industrialized sources (e.g., metals);
- over long periods (decades); and
- to particularly sensitive plant species (e.g., swiss chard).

Yields of tolerant crops may actually benefit from trace element levels in soils that could drastically reduce yields of more sensitive crops (Chapman, 1966).

5.3.1 Crop yields

The farmer substituting sludge in whole or in part for commercial NPK fertilizer is interested primarily in maximizing his yield of marketable products. If sludge is applied to land at no cost to him, he can accept lower yields than those obtained from NPK fertilizer, up to the equivalent cost of the commercial fertilizer itself. Yields equal to or greater than those obtained from NPK fertilizer would, in the short run, be sufficient justification for switching from commercial fertilizer to sludges.

Table 11 summarizes the cumulative yield of plant material from both lysimeter experiments, up to the end of the 1976 growing season.

The information of major importance from this table for the fluid sludge-orchard grass system, is:

1. To obtain orchard grass yields from field sludge application (regardless of source) equivalent to NPK yields, at least 200% of the NPK nitrogen rates must be applied.
2. Highest grass yields were obtained at all application rates with iron sludge on both soils. At the highest iron sludge rate (5100 kg TKN/ha) yields exceeded NPK fertilizer yields by 38% on loamy sand and 28% on silt loam.
3. Equal sludge TKN applications, at the highest rate, yielded approximately 25% more orchard grass on silt loam than on loamy sand for all three sludge sources.

TABLE 11. CUMULATIVE YIELD OF PLANT DRY MATTER (tons/ha) FROM THE FLUID SLUDGE-ORCHARD GRASS (1973 to 1976) AND AIR-DRIED SLUDGE-WHEAT (1974 to 1976) EXPERIMENTS

* Treatment (kg TKN/ha)	Orchard Grass		** Treatment (kg TKN/ha)	Wheat/Buckwheat	
	Loamy Sand	Silt Loam		Sand	Clay
NPK (1700)	50	67	NPK (300)	28	35
Alum (1700	31	47	Alum (1186)	23	35
(3400)	49	63	(2372)	24	36
(5100)	60	75	(3558)	25	37
Iron (1700)	36	49	Iron (829)	20	34
(3400)	51	65	(1658)	21	35
(5100)	69	86	(2487)	22	36
Lime (1700)	32	45	Lime (2328)	19	35
(3400)	48	61	(4656)	24	35
(5100)	59	74	(6984)	27	34

* Fluid sludge rates based on TKN.

** Air-dried sludge rates calculated for TKN from zinc equivalent rates.

Figure 2 (Annual Orchard Grass Yields 1973 to 1976) shows the increasing effect of residual organic nitrogen mineralization with time. During the first crop year (1973), only two out of nine sludge fertility treatments gave yields equal to or better than NPK on loamy sand. By the fourth year (1976), eight out of nine sludge treatments produced yields in excess of the annual NPK yield. While the benefit of sludge treatment was less pronounced on the more productive silt loam soil, the number of sludge treatments equal to or better than NPK yields rose from three out of nine in 1973 to five out of nine in 1976.

Figure 3 (Annual Crop Yields 1974 to 1976) from the air-dried sludge lysimeter experiment shows a much different picture than did the fluid sludge-orchard grass experiment. The dramatic yield increases with increasing rates of sludge application do not occur on either sand or clay soils receiving air-dried sludges. The sand soil, in particular,

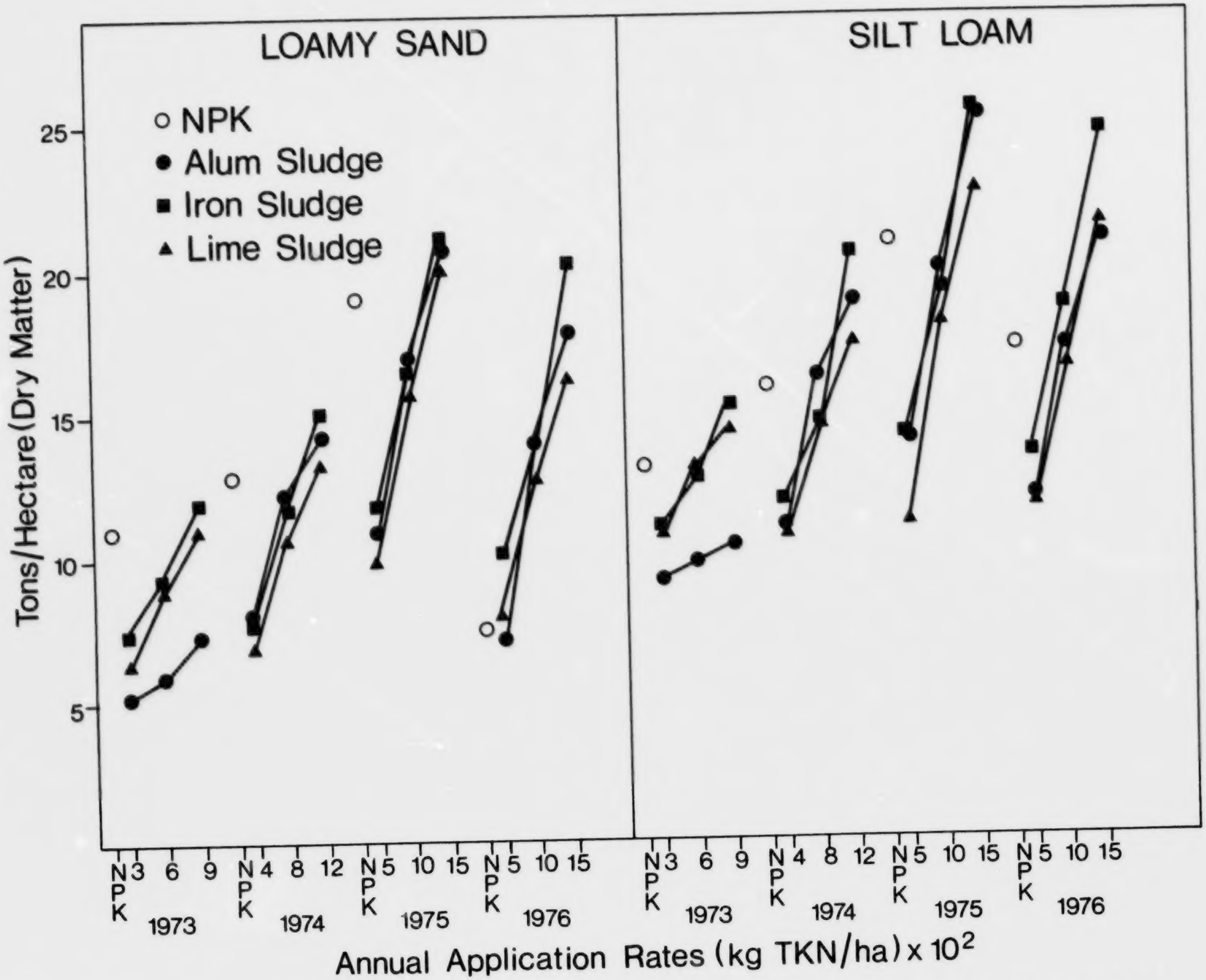


Figure 2 ANNUAL ORCHARD GRASS YIELDS (1973-76) FLUID SLUDGE LYSIMETER EXPERIMENT

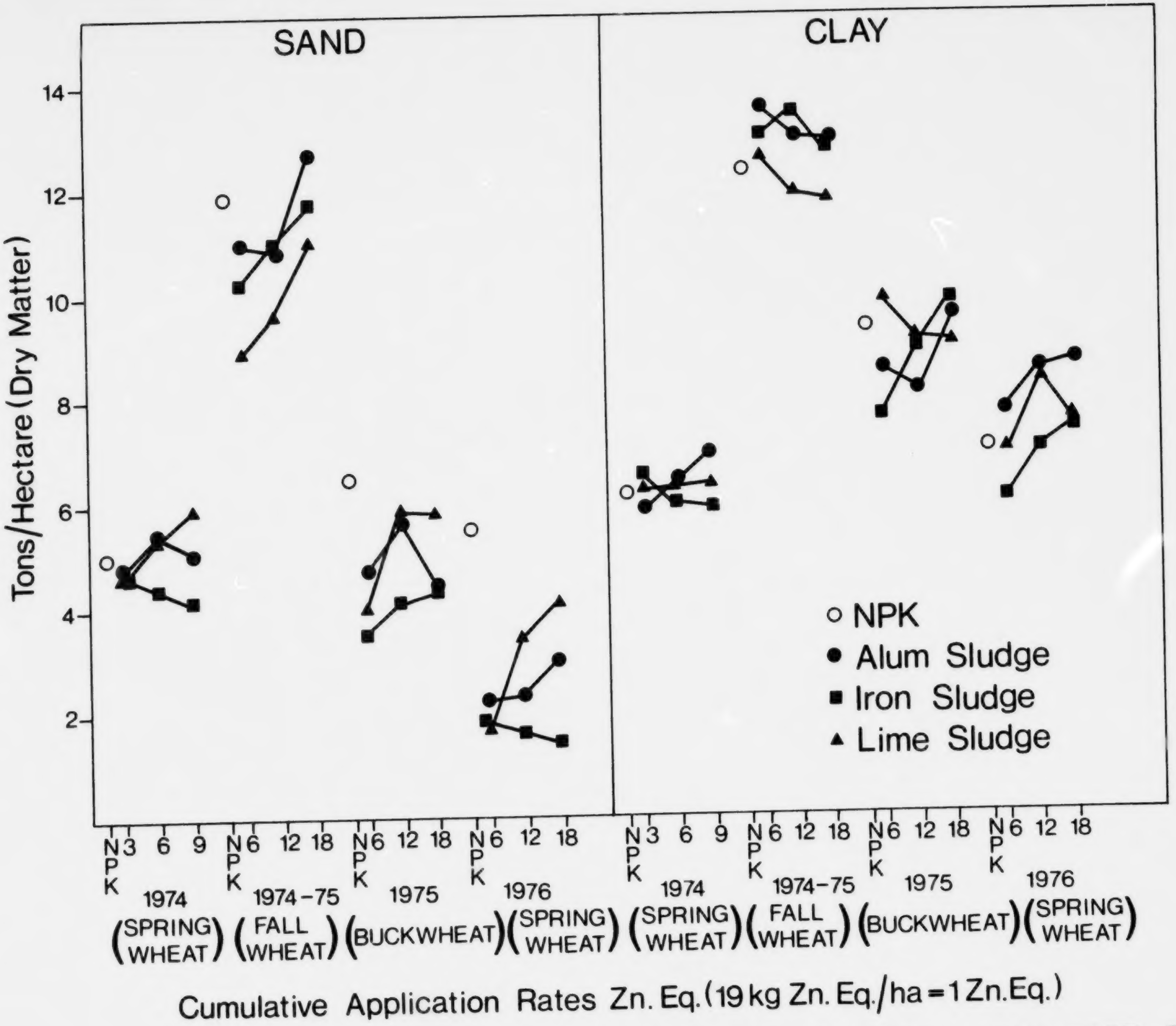


Figure 3 ANNUAL CROP YIELDS (1974-76) AIR-DRIED SLUDGE LYSIMETER EXPERIMENT

does not show any beneficial effect of residual organic nitrogen mineralization with time. During the first crop year, seven out of nine sludge treatments produced yields approximately equal to NPK on sand. By the fourth crop harvest in 1976, none of the sludge treatments produced yields equal to NPK. Increased loadings of iron sludge produced a slightly negative crop yield response. The 1976 NPK yield of spring wheat on sandy soil was, however, greater than the 1974 NPK yield. Lack of soluble nitrogen in the required amount at the appropriate time is, therefore, assumed to be a major factor limiting yields in the air-dried sludge applied to sandy soil.

Cumulative yield response from the more productive clay soil to sludge treatments was equivalent to NPK fertilizer yields. No significant sludge rate effect was evident, indicating that the rate of organic nitrogen mineralization in the air-dried sludge-soil mixture was not proportional to TKN sludge loadings. Fall wheat yields were approximately double spring wheat yields for all treatments on both soils. Fall wheat is better adapted than spring wheat to climatic conditions in southern Ontario.

Figures 4 and 5 show the cumulative yields of orchard grass and wheat crops, respectively. These figures show that:

- Fluid sludges can supply both immediately available and residual nitrogen to forage crops (e.g., orchard grass) requiring relatively large amounts of soluble N during the growing season.
- Air-dried sludges having less than 2% of TKN in soluble form must be supplemented with an NH_4-N type of fertilizer if economic yields of cereals (e.g., wheat) are to be maintained over many years, particularly on sandy soils.
- Yields tend to increase with increasing cation-exchange capacity of soils using either sludge or NPK fertilizers at equivalent rates.
- Iron sludge at the highest rate produced approximately 15% more yield of orchard grass on both soils than did alum or lime sludges at the same rate.

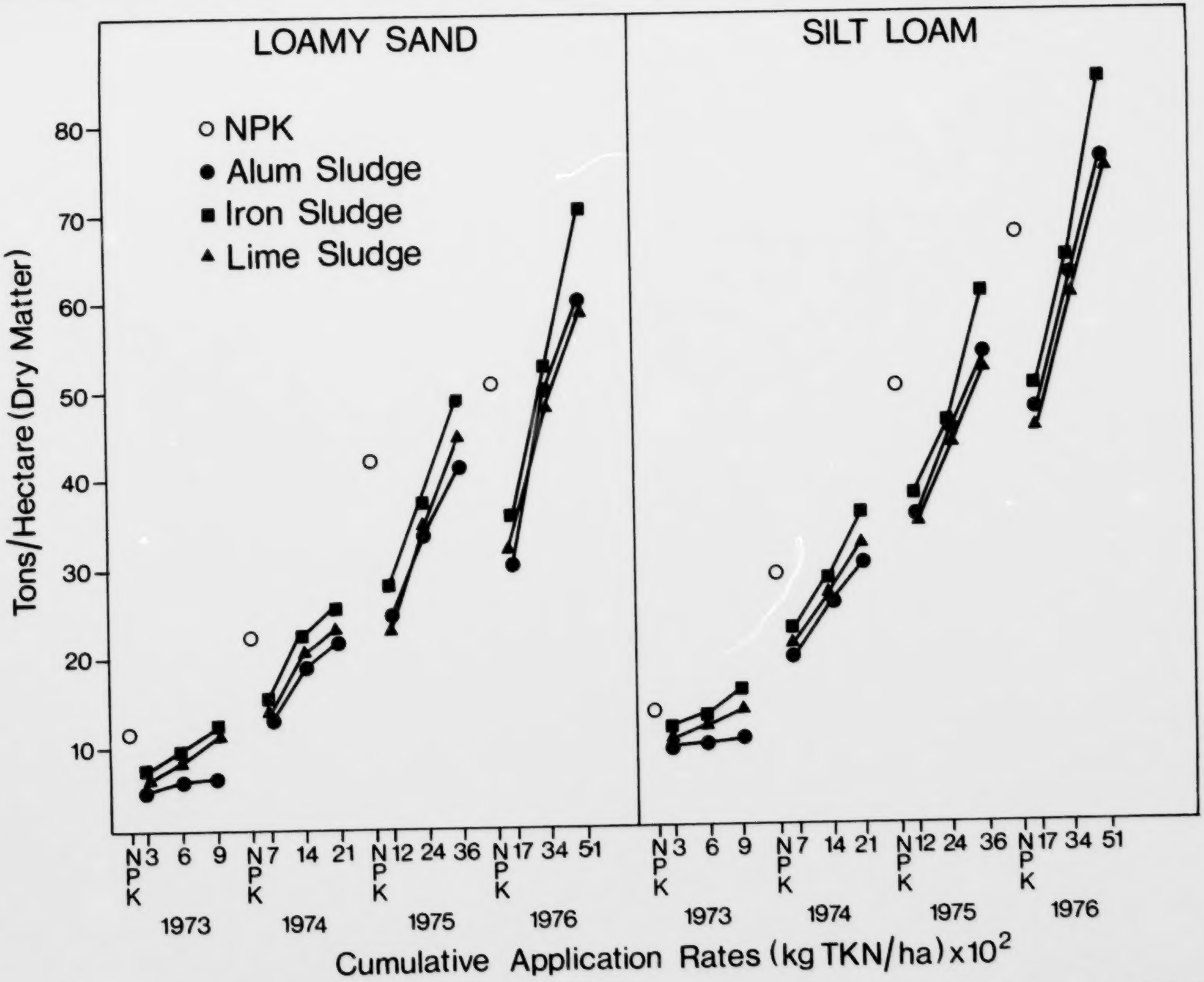


Figure 4 CUMULATIVE ORCHARD GRASS YIELDS (1973-76) FLUID SLUDGE LYSIMETER EXPERIMENT

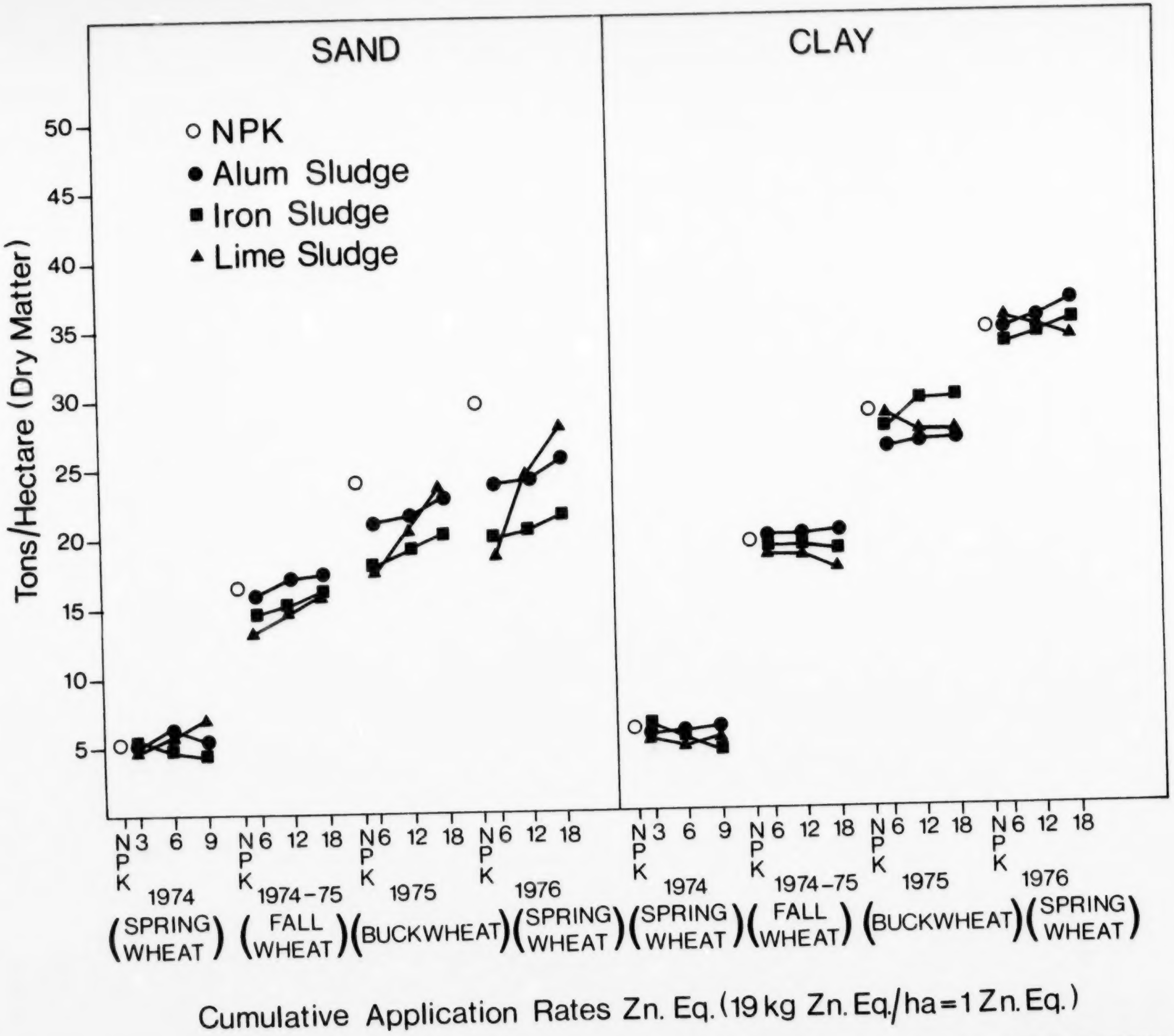


Figure 5 CUMULATIVE CROP YIELDS (1974-76) AIR-DRIED SLUDGE LYSIMETER EXPERIMENT

Total yields of plant dry matter from air-dried sludge applications to sand and clay soils do not reflect the relative yields of straw and the more economically valuable grain. Table 12 summarizes wheat grain yields for the wheat crops harvested during 1974, 1975, and 1976.

The cumulative wheat grain yield from all sludge treatments equalled 74% of the NPK cumulative grain yield on sandy soil and 97% of the NPK grain yield on clay. The relative yield of grain from the most recent (spring 1976) crop declined to only 38% of NPK yield on sandy soil, but increased to 103% of NPK yield on clay soil. Increased lime sludge application rates resulted in increased grain yield for sandy soils. However, this effect was not observed for clay soils.

5.3.2 Plant chemical constituents

The constituents of concern with regard to quality of plants grown on sludge amended soils are the micronutrient heavy metals, some of which are potentially toxic to plants and/or animals, and the macronutrients (N, P, K).

Table 13 summarizes the potential toxicity of the heavy metals Cd, Cr, Cu, Ni, Pb, and Zn to crops grown on sludged soils, to animals fed crops grown on contaminated soil, or to animals fed directly without intermediary crop uptake (Ryan, 1976). Other constituents of concern (e.g., Hg, Mo, Se, Co) have not been analyzed long enough for them to be included in this discussion.

Plant tissue analysis is generally regarded as the most appropriate evaluator of heavy metal buildup trends before irreparable damage results (CAST, 1976). Analysis of standard portions of plant tissue at regular intervals should indicate whether metal availability to plants is increasing to levels requiring corrective measures or cessation of sludge application.

TABLE 12. WHEAT GRAIN YIELDS (tons/ha) AIR-DRIED SLUDGE EXPERIMENT (1974 to 1976)

Treatment/Rate	Sand				Clay			
	Spring (74)	Fall (75)	Spring (76)	Total (74-76)	Spring (74)	Fall (75)	Spring (76)	Total (74-76)
NPK	1.42	4.71	2.74	8.87	1.68	5.78	3.31	10.77
Alum (low)	1.26	4.53	1.02	6.81	1.50	5.79	3.40	10.69
(medium)	1.31	4.12	0.92	6.35	1.42	5.17	3.74	10.33
(high)	1.15	4.87	1.34	7.36	1.65	5.31	3.67	10.63
Iron (low)	1.23	4.45	0.74	6.42	2.03	5.88	2.97	10.88
(medium)	1.34	4.36	0.72	6.42	1.71	5.70	3.44	10.85
(high)	1.11	4.79	0.64	6.54	1.41	5.35	3.54	10.30
Lime (low)	0.98	3.65	0.62	5.25	1.61	5.64	3.38	10.63
(medium)	1.30	3.49	1.66	6.45	1.65	5.05	3.61	10.31
(high)	1.59	4.36	1.74	7.69	1.68	4.86	2.97	9.51
Mean of all sludge treatments $(\bar{x})$	1.25	4.29	1.04	6.58	1.63	5.42	3.41	10.46
Ratio $(\bar{x})$/NPK	0.88	0.91	0.38	0.74	0.97	0.94	1.03	0.97

TABLE 13. POTENTIAL TOXICITY OF HEAVY METALS TO PLANTS AND ANIMALS (Ryan, 1976)

Element	Essential for Growth		Toxicity		
	Plant	Animal	Plant[1]	Animal[2]	Animal[3]
Cd	-	-	Moderate	High	High
Cr	-	Yes	Low	Low	Low
Cu	Yes	Yes	High	Moderate	Slight
Ni	-	Yes	High	Moderate	Low
Pb	-	-	Low	High	Low
Zn	Yes	Yes	Moderate	Low	Low

[1] Metal applied to soil on which crop is grown.
[2] Metal fed directly to animal.
[3] Animal fed crop grown on sludged soil.

Some of the questions which plant tissue analysis of crops grown on sludge amended soils attempts to answer include the following:

1. Does the rate of sludge application influence the rate of heavy metal uptake on both a short term and long term basis? In other words, do plant "constituents of concern" increase in concentration with each successive sludge application?
2. Does cessation of sludge application (i.e., a single heavy dose followed by several years without sludge application) make metals more or less available to plants grown on these sludge amended soils?
3. Are there significant differences in plant metal uptakes related to differences in soil characteristics (e.g., pH <6.0)?
4. Are there any significant differences between sludge types (e.g., alum, iron and lime chemical sludges) with regard to uptake or concentration of specific metals?

5. Do the plant metal concentrations obtained from sludge treatments differ significantly from those from the "normal" agronomic practice represented by the NPK control treatment?
6. How do the concentrations of the various "constituents of concern" compare with the normal concentration ranges reported in the literature?
7. Which, if any, sludge treatments result in plant metal concentrations which approach or exceed suggested maximum allowable "tolerance" or "toxic" concentrations mentioned in the literature?
8. How important are plant species or variety differences in deciding whether a particular metal concentration is tolerable (e.g., in accumulator species) or dangerous to the health of the plant or its consumers?

Although results of these lysimeter experiments cannot adequately answer all of the questions posed or be loosely applied to all other sludge-soil plant situations, certain trends that have developed over sufficient time are of general interest.

Tables 14, 15, 16 and 17, summarize the heavy metals quality (μg/g dry matter) of plant tissue from both sludge treatments and controls compared to suggested "tolerance" and "toxic" concentrations for orchard grass, wheat grain, wheat straw and buckwheat, respectively. The sludge treatment values include both the mean of all nine sludge treatments as well as the maximum average annual value from any sludge treatment. A comparison of the mean and maximum values indicates the extent to which sludge loading rates influence plant metals concentrations.

For the 14 cuts of orchard grass analyzed between 1973 and 1976 (Table 14), none of the six metals exceeds suggested tolerance or toxic levels, with the exception of lead (maximum value) on loamy sand. The very high lead values obtained from the NPK controls on both soils (8.0 to 8.8 μg/g) suggest that lead emission from automobiles and the close proximity of the Burlington-Skyway bridge to the location of the lysimeter experiments may account for this unusual result. Other investigators also have shown lead adsorption (even on washed plants) to be directly related to proximity to major highways.

TABLE 14. ORCHARD GRASS QUALITY FROM SLUDGE AMENDED SOILS AND CONTROL SOILS (1973 to 1976) COMPARED TO SUGGESTED TOLERANCE AND TOXIC LEVELS FOR HEAVY METALS (μg/g dry matter)

Constituent	NPK Control		Sludge Amended Soils				Suggested Tolerance Levels (a)	Suggested Toxic Levels (b)
			Mean of All Treatments		Maximum Average Annual Values			
	Loamy Sand	Silt Loam	Loamy Sand	Silt Loam	Loamy Sand	Silt Loam		
Cadmium	0.24	0.23	0.24	0.24	0.29	0.30	3.0	
Copper	6.4	6.6	8.1	7.3	10.5	9.6	150	>20
Chromium	1.1	1.0	1.2	1.1	1.2	1.2	2.0	
Lead	8.0	8.8	8.4	7.6	11.4	9.5	10.0	
Nickel	1.1	1.2	1.3	1.3	1.4	1.3	3.0	>50
Zinc	48	45	59	48	84	64	300	>200

(a) Source: Soil-Plant Relationships, (Melsted, 1973).
(b) Source: Fate and Effects of Trace Elements in Sewage Sludge When Applied to Agricultural Lands, (Page, 1974).

TABLE 15. WHEAT GRAIN QUALITY FROM SLUDGE AMENDED SOILS AND CONTROL SOILS (1974 to 1975) COMPARED TO SUGGESTED TOLERANCE AND TOXIC LEVELS FOR HEAVY METALS (μg/g dry matter)

Constituent	NPK Control		Sludge Amended Soils				Suggested Tolerance Levels (a)	Suggested Toxic Levels (b)
			Mean of All Treatments		Maximum Average Annual Values			
	Sand	Clay	Sand	Clay	Sand	Clay		
Cadmium	0.16	0.11	0.19	0.14	0.20	0.15	3.0	
Copper	6.4	10.5	9.0	7.9	9.4	8.3	150	>20
Chromium	1.2	1.3	1.4	1.4	2.1	1.8	2.0	
Lead	0.9	0.7	0.6	0.7	1.0	1.0	10.0	
Nickel	0.8	1.1	0.7	0.8	0.7	0.9	3.0	>50
Zinc	62	47	50	51	54	55	300	>200

(a) Source: Soil-Plant Relationships, (Melsted, 1973).
(b) Source: Fate and Effects of Trace Elements in Sewage Sludge When Applied to Agricultural Lands, (Page, 1974).

TABLE 16. WHEAT STRAW QUALITY FROM SLUDGE AMENDED SOILS AND CONTROL SOILS (1974 to 1975) COMPARED TO SUGGESTED TOLERANCE AND TOXIC LEVELS FOR HEAVY METALS (µg/g dry matter)

Constituent	NPK Control		Sludge Amended Soils				Suggested Tolerance Levels (a)	Suggested Toxic Levels (b)
			Mean of All Treatments		Maximum Average Annual Values			
	Sand	Clay	Sand	Clay	Sand	Clay		
Cadmium	0.24	0.19	0.19	0.19	0.22	0.20	3.0	
Copper	3.0	2.9	4.0	4.2	4.7	4.8	150	>20
Chromium	0.85	0.90	0.85	0.90	0.90	0.90	2.0	
Lead	8.1	5.7	6.0	6.7	7.8	7.3	10.0	
Nickel	1.3	2.0	1.6	1.3	2.0	3.0	3.0	>50
Zinc	24	18	26	20	30	23	300	>200

(a) Source: Soil-Plant Relationships, (Melsted, 1973).
(b) Source: Fate and Effects of Trace Elements in Sewage Sludge When Applied to Agricultural Lands, (Page, 1974).

TABLE 17. BUCKWHEAT (GREEN CROP) QUALITY FROM SLUDGE AMENDED SOILS AND CONTROL SOILS COMPARED TO SUGGESTED TOLERANCE AND TOXIC LEVELS FOR HEAVY METALS (μg/g dry matter)

Constituent	NPK Control		Sludge Amended Soils				Suggested Tolerance Levels (a)	Suggested Toxic Levels (b)
			Mean of All Treatments		Maximum Average Annual Values			
	Sand	Clay	Sand	Clay	Sand	Clay		
Cadmium	0.90	0.25	0.48	0.30	0.56	0.47	3.0	
Copper	6.3	7.8	8.8	9.8	9.7	13.2	150	>20
Chromium	0.70	0.70	0.70	0.70	0.80	0.80	2.0	
Lead	11.6	8.6	11.1	9.2	12.0	10.3	10.0	
Nickel	1.4	1.5	0.9	1.2	1.3	1.3	3.0	>50
Zinc	58	28	57	40	69	47	300	>200

(a) Source: Soil-Plant Relationships, (Melsted, 1973).
(b) Source: Fate and Effects of Trace Elements in Sewage Sludge When Applied to Agricultural Lands, (Page, 1974).

Table 15 summarizes the metals concentrations in the two wheat grain crops harvested in 1974 and 1975. As indicated in the literature (Kirkham, 1976), grains tend to exclude heavy metals more than do foliage or leafy portions of crops. With the exception of chromium, the maximum metal concentrations in wheat grain are lower than the maximum values observed in orchard grass. The order of magnitude of reduction in wheat grain lead concentration compared to orchard grass is particularly noteworthy. The grain is physically protected from atmospheric contamination to a greater degree than is foliage. These results lend credence to the hypothesis that the high lead values observed in Table 14 were not a result of plant uptake of the not readily available lead from the sludge treated soils. The maximum value for chromium (2.1 μg/g) obtained when the high rate of iron sludge was applied to sandy soil slightly exceeds the suggested "tolerance" level of 2.0 μg/g (Melsted, 1973); however, increased chromium in the food chain may be desirable due to a general deficiency in the human diet (CAST, 1976). Of all six heavy metals monitored, chromium is of least concern with regard to either plant or animal toxicity (Table 13).

Table 16 summarizes the metal concentrations in the two wheat straw crops harvested in 1974 and 1975. With the exception of nickel, which approaches the maximum suggested tolerance level of 3.0 μg/g when iron sludge at the highest rate is applied to clay soil, the concentration of all other metals is acceptable. The mean and maximum values of heavy metals in wheat straw from sludge treatment of clay and sandy soils are similar to values obtained for orchard grass, with the exception of copper and zinc concentrations which are significantly lower in wheat straw.

The buckwheat crop (Table 17) was harvested as a green crop (prior to maturity) and can, therefore, be compared to the orchard grass and wheat straw values. As was the case with orchard grass, lead concentrations exceed the maximum suggested "tolerance" level of 10.0 μg/g for sludge treatments on both soils and NPK treatment on sandy soil. Cadmium is the only metal whose average and maximum concentrations in buckwheat plant tissue are significantly greater than those found in orchard grass or wheat straw. The highest cadmium value (0.56 μg/g) was, however, well below the suggested maximum tolerance level of 3.0 μg/g and significantly lower than the cadmium concentration from the NPK control on sandy soil (0.9 μg/g). Despite the low pH (5.6) of the sandy soil, none of the heavy metal concentrations in plants grown on this soil for three consecutive years, indicated any long term trends which could be considered cause for concern.

A word of caution is in order to those trying to predict potentially hazardous trends from a limited data base. Most of the results reported in the literature are based on greenhouse pot studies rather than large scale lysimeter or field studies. Even when field experiments are compared, conflicting trends are frequently reported. For example, Decker et al (1975) have reported that applications of 2 kg Cd/ha have caused a fivefold increase in the Cd content of corn grain from 0.12 to 0.68 ppm. Kelling et al (1976) found that application of 4.3 kg Cd/ha did not cause any significant change in Cd content of corn grain. The resolution of these conflicting results is difficult and this indicates the need to define each separate system on some common basis to allow for meaningful comparison.

5.3.2.1 Heavy metals

Cadmium. The World Health Organization (WHO) has recommended that adult humans should consume not more than 500 μg Cd/week (WHO, 1972). Linnman et al (1973) suggest that Cd intake from wheat grain grown on sludged soil could increase beyond the level recommended as tolerable by WHO. Investigations in the U.S.A. (CAST, 1976) with repeated annual sludge applications to soil cropped to corn, indicated that amounts applied in any given year had a much greater influence on the cadmium content of the plant than did cumulative amounts applied. The implication of these results is that most of the cadmium applied was being converted, over time, to forms of relatively low availability to plants. Similar observations have been made by Sidie and Kardos (1976) with regard to concentrations of Cd, Cu and Zn, in reed canary grass and corn silage.

Figure 6 shows a decline, with time, in orchard grass cadmium (loamy sand) from 0.54 μg/g in 1973 (mean of all sludge treatments) to 0.12 μg/g in 1976. A similar trend is evident for orchard grass grown

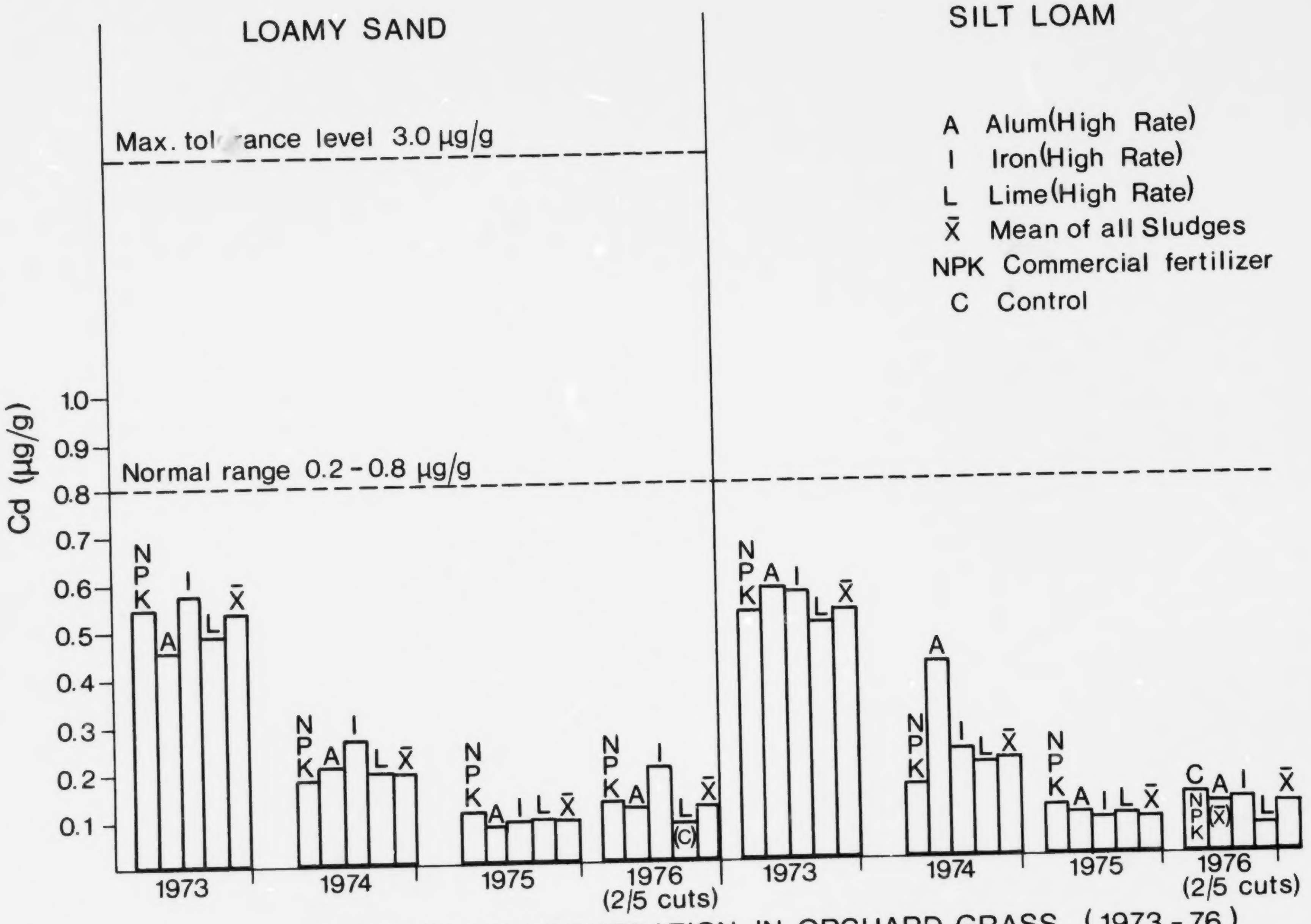


Figure 6 CADMIUM CONCENTRATION IN ORCHARD GRASS (1973 - 76)

on silt loam. Figure 7 shows a decline in wheat grain cadmium concentration (sandy soil) from 0.28 μg/g for 1974 spring wheat to 0.10 μg/g for 1975 fall wheat. A similar trend is evident for wheat grain grown on clay soil.

NPK fertilizer applications may increase cadmium uptake to a greater extent than do sludge treatments (CAST 1976). This is shown in Figures 8 and 9 where cadmium concentration deviations from NPK controls (average annual values) are given for orchard grass and wheat grain and straw, respectively. If one accepts NPK fertilizer application as "standard agronomic practice", then deviations of Cd from this standard for all sludge treatments to all soils and crops investigated were well within the "normal" range of variability. Cadmium, the metal constituent of greatest concern to human health did not, therefore, show any cumulative trends of concern under the conditions of these lysimeter experiments.

Copper. Keeney and Walsh (1975) found that the extractable copper content of sludge treated soil decreased with time, suggesting that a reversion to less soluble forms was occurring. Copper concentrations in excess of 20 ppm in the leafy portions of plants have been associated with plant toxicity (Walsh, 1972). Copper has been added to soils deficient in this necessary micronutrient. Chickens and pigs fed a copper rich diet show better weight gain than control animals (CAST, 1976).

Copper concentrations in orchard grass grown on loamy sand (mean of all sludge treatments) increased slightly with time, averaging 7.8 μg/g in 1973 and 9.1 μg/g in 1976. Unlike cadmium, all sludge treatments tended to increase the copper concentration deviation from NPK control (Figure 10). The high rate application of iron sludge on loamy sand mean of 14 cuts (1973 to 1976) showed the highest deviation (+4.2 μg/g), while lime sludge on silt loam showed the lowest deviation from NPK control (+0.8 μg/g). Copper concentrations in wheat grain grown on clay soil were 2 to 4 μg/g less than NPK control values (Figure 11).

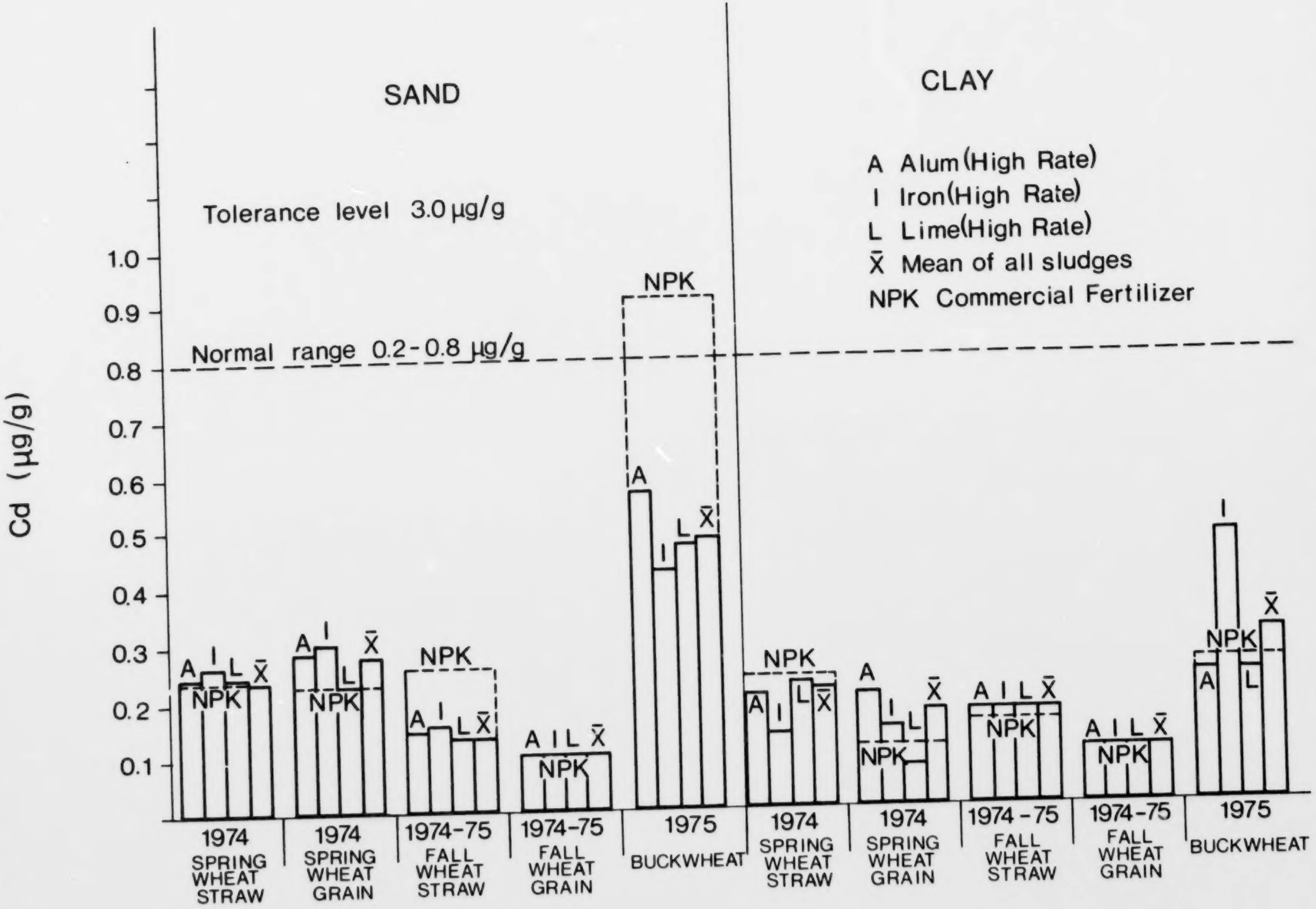


Figure 7 CADMIUM CONCENTRATION IN PLANT TISSUE (GRAIN/STRAW) (1974-75)

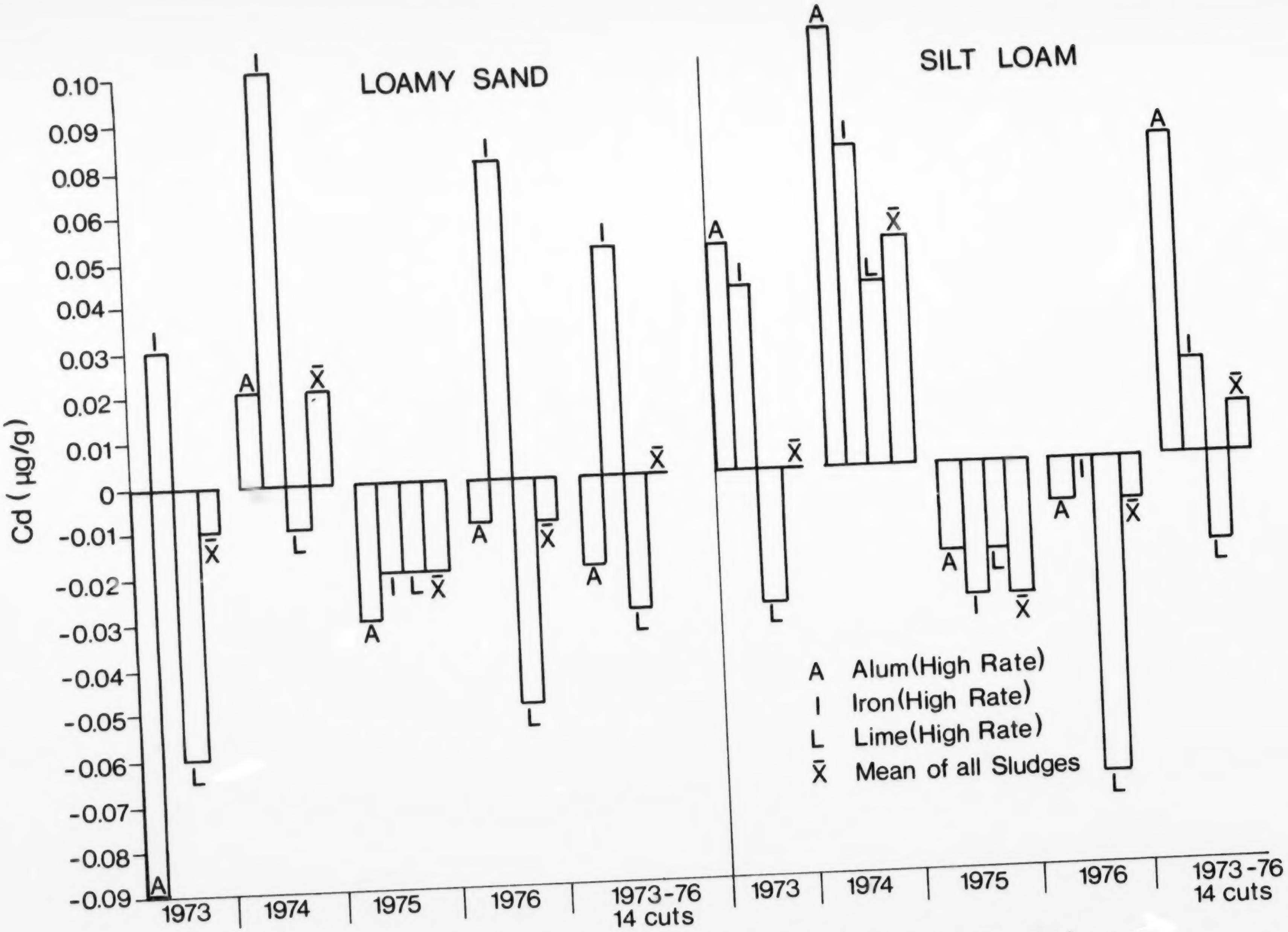


Figure 8

CADMIUM CONCENTRATION IN ORCHARD GRASS (1973 - 76)

(Deviation From NPK Control)

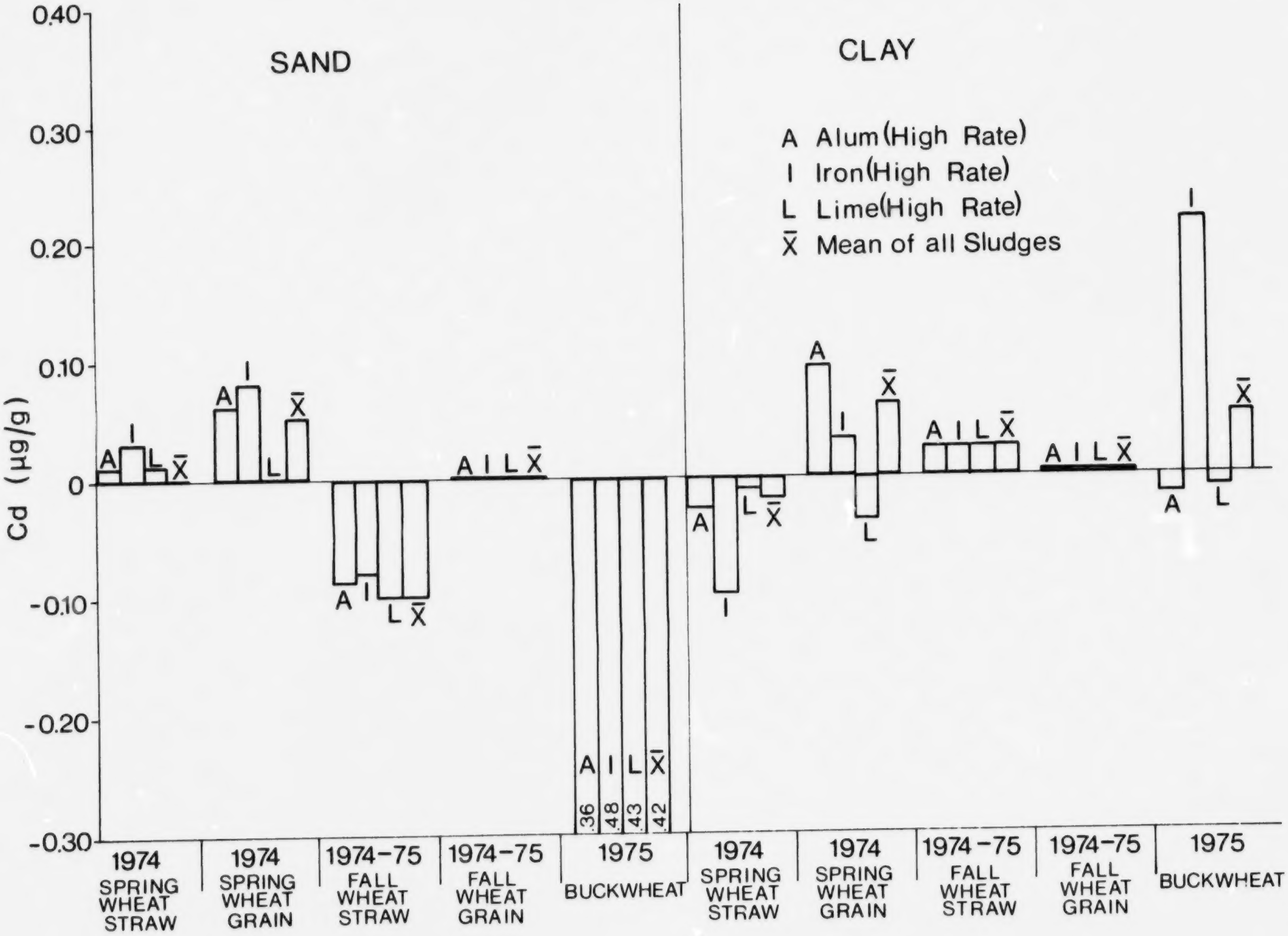


Figure 9 CADMIUM CONCENTRATION IN PLANT TISSUE (GRAIN/STRAW) (1974 - 75)

(Deviation From NPK Control)

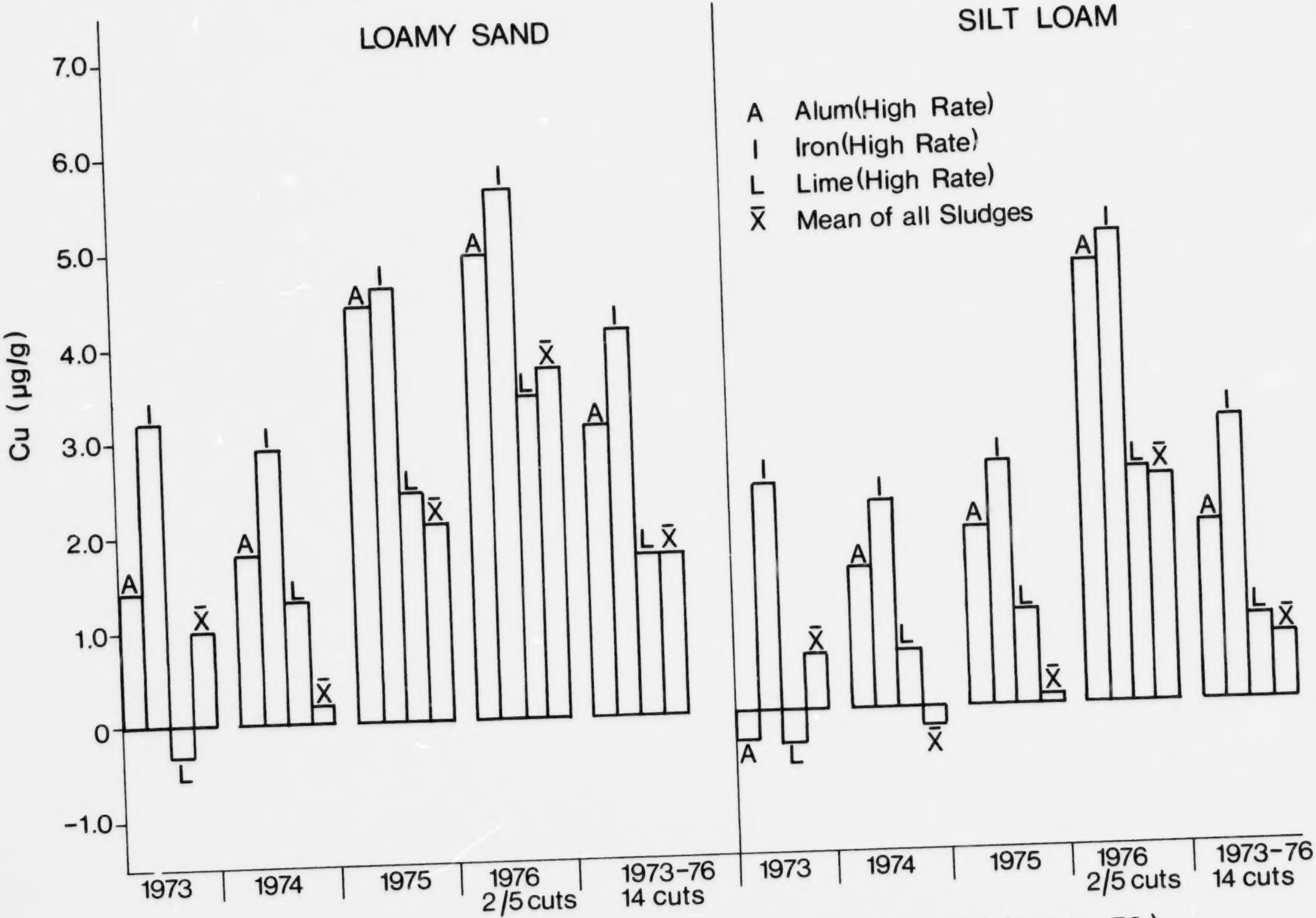


Figure 10 COPPER CONCENTRATION IN ORCHARD GRASS (1973 - 76)

(Deviation From NPK Control)

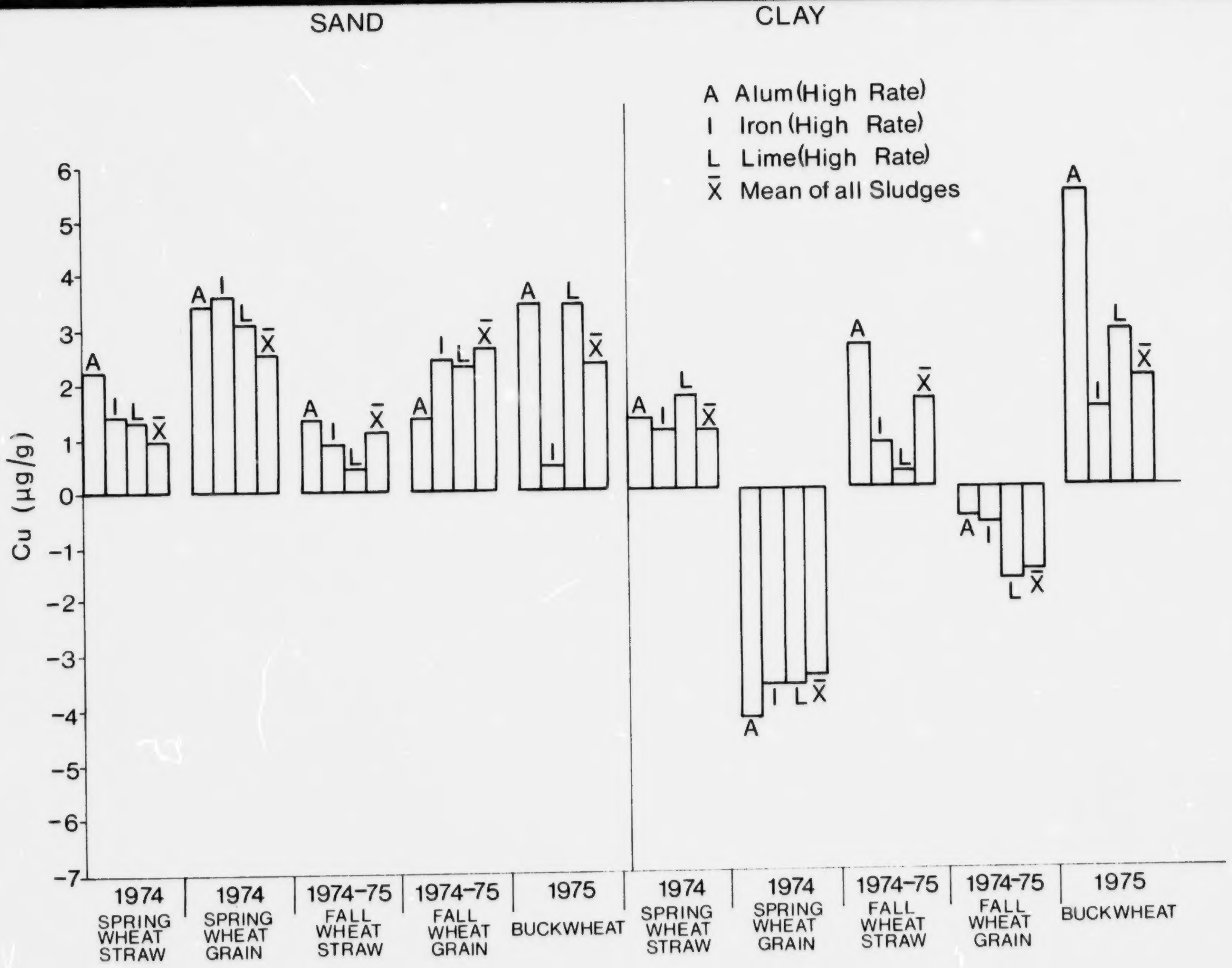


Figure 11 COPPER CONCENTRATION IN PLANT TISSUE (GRAIN/STRAW) (1974-75)

(Deviation From NPK Control)

Zinc. Zinc toxicity may occur if the leafy tissues of most agricultural crops contain in excess of 200 µg/g (Page, 1974). In some instances, a moderate increase in the zinc content of the food supply may be beneficial because there is evidence that human diets are often deficient in zinc. Zinc concentrations in orchard grass reached a maximum value of 112 µg/g in 1974 at the high rate of iron sludge addition to loamy sand. Despite additional sludge applications, however, zinc concentrations declined in 1975 and 1976 for all treatments with the highest value recorded in 1976 (78 µg/g) from iron sludge at the highest rate (Figure 12). Deviations from NPK control did not increase with successive sludge applications to orchard grass. Similarly, application of dried sludge to sand and clay soils showed no trend with time towards increased zinc concentrations in wheat straw, wheat grain or buckwheat. The concentration of zinc in the wheat grain crop grown on sand (mean of all sludge treatments) was significantly lower (Table 15) than the corresponding zinc concentration from the NPK control.

Nickel. Nickel is toxic to plants at concentrations above 50 ppm in plant tissue (Cunningham et al, 1975). Toxicity has thus far been reported only on acid soils. The suggested maximum "tolerance" concentration of 3.0 µg/g (Melsted, 1973) may, therefore, be conservative for neutral or alkaline soils. Nickel may be required by chickens and domestic animals as a supplement to their normal dietary intake (CAST, 1976).

Nickel in spring wheat exceeded the suggested maximum tolerance level at the high rate of application of iron sludge on sand, and the high rate of lime sludge on clay. Subsequent crops of fall wheat (straw and grain) and buckwheat on both soils showed lower values than those observed in the first wheat straw crop for all treatments (Figure 13). Nickel concentrations in orchard grass remained relatively constant over four years, never exceeding 1.8 µg/g. Deviations from NPK control in orchard grass and wheat tissues varied from year to year without any definite trend towards nickel buildup.

Chromium. Hexavalent chromium is toxic to plants but anaerobic digestion of sludges reduces the hexavalent form to trivalent chromium which is relatively insoluble and unavailable to plants. Although some

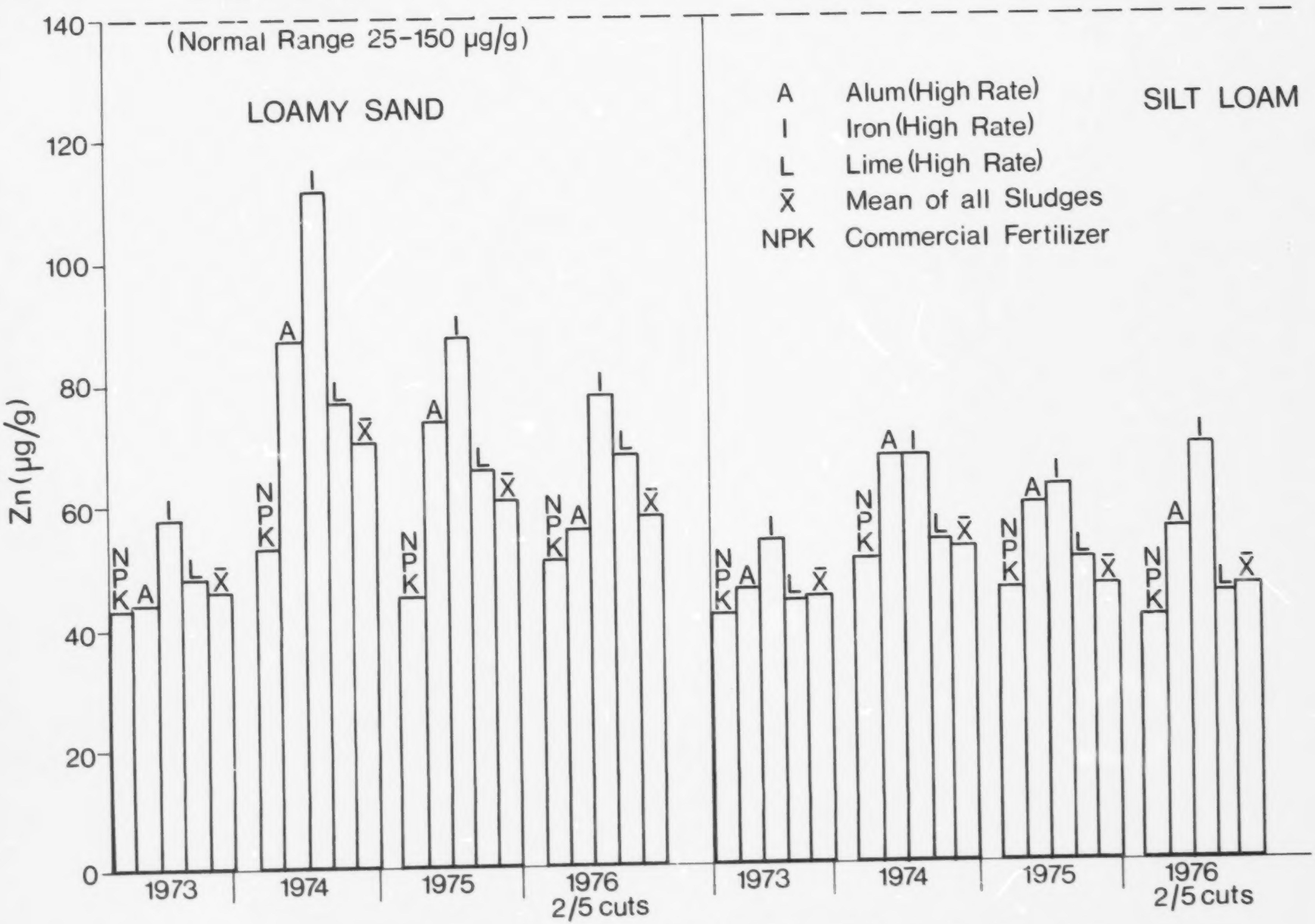


Figure 12 ZINC CONCENTRATION IN ORCHARD GRASS (1973-76)

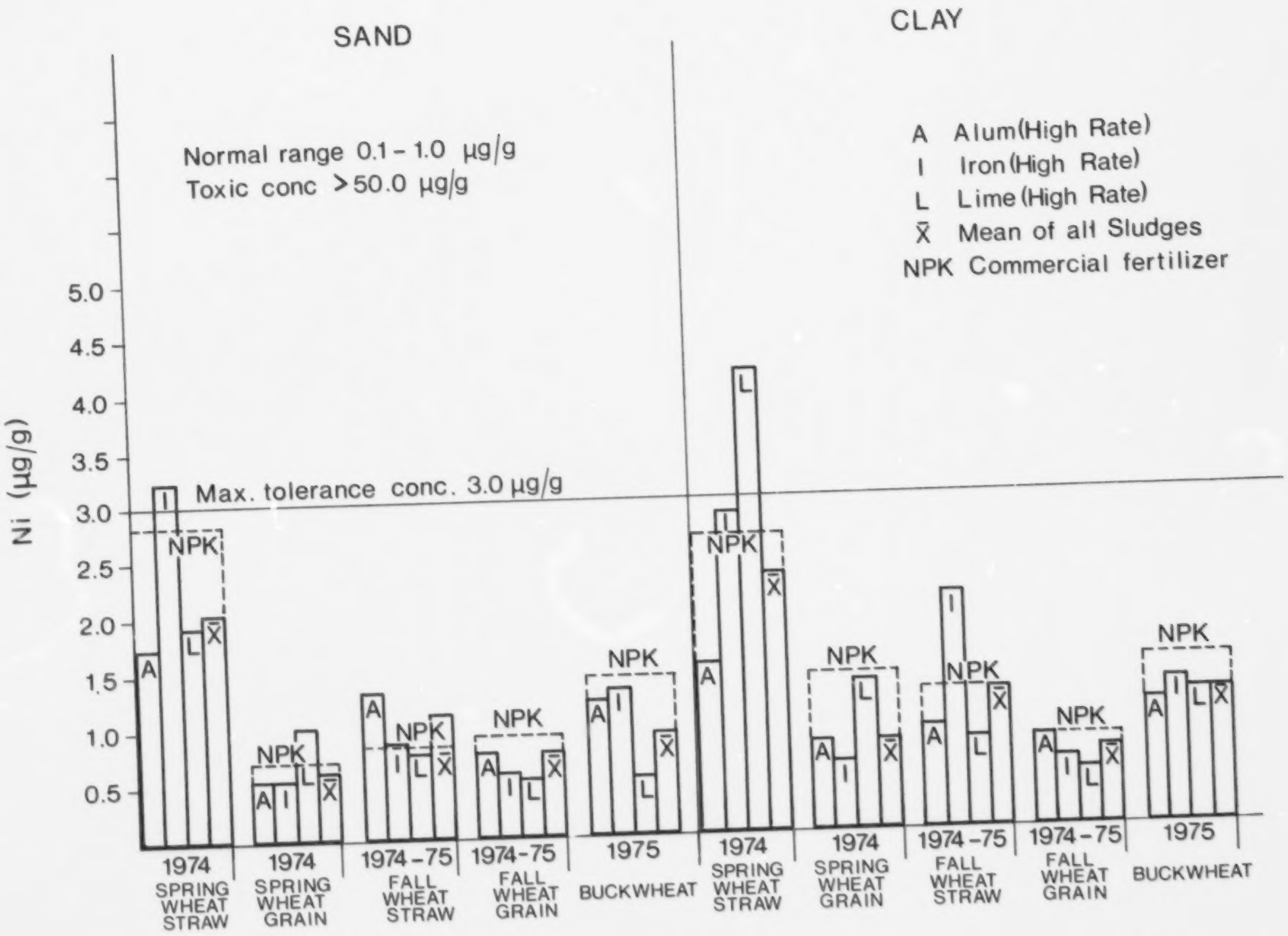


Figure 13 NICKEL CONCENTRATION IN PLANT TISSUE (GRAIN/STRAW) (1974 - 75)

plant species normally absorb up to 10 μg/g, the maximum tolerance level suggested by Melsted (1973) is 2.0 μg/g. Clapp et al (1976), found no increase in chromium concentration above the baseline level of 4 μg/g in corn leaf tissue when up to 530 kg Cr/ha was applied with sewage sludge. Concentration in the corn grain never exceeded 0.1 μg/g. There have been several reports of slight yield increases following chromium additions to soil. There is evidence that this metal is required by humans and animals and that diets may be deficient in chromium in certain areas (Underwood, 1971).

Orchard grass chromium concentrations exceeded the maximum suggested tolerance level of 2.0 μg/g (1976 only) on both soils. The highest concentration observed was 2.4 μg/g from the high rate alum sludge addition to loamy sand. Fall wheat grain (1975) chromium concentrations were significantly higher than the spring wheat grain concentration of 0.7 μg/g, and exceeded the suggested "tolerance" level at the highest rate of alum and iron sludge additions to both soils (Figure 14). In view of the previous discussion, these concentrations are not considered to be a cause for concern at this time.

Lead. Lead exhibits a low degree of potential toxicity to plants and a high degree of potential toxicity to animals. Hassett (1974) found the lead sorption capacity of Illinois soils to be several thousand kg/ha, increasing as the pH, CEC and extractable phosphorus content of the soil increase. Baumhardt and Welch (1972) concluded that the lead content of soils would have to exceed 1% and the soil pH would have to be below 5.5 before significant deleterious effects on plant growth would be encountered.

Although the concentrations of lead in orchard grass were unusually high, exceeding the suggested maximum tolerance concentration of 10 μg/g on both soils in 1974 and 1976, the (1973 to 1976) deviation from NPK control was quite small, averaging only +0.4 μg/g on loamy sand and -1.2 μg/g on silt loam. Similarly, concentrations of lead in wheat and buckwheat occasionally approached or exceeded 10 μg/g but the deviation from NPK control was frequently zero or negative (Figure 15). This suggests that the source of lead contamination was air pollution

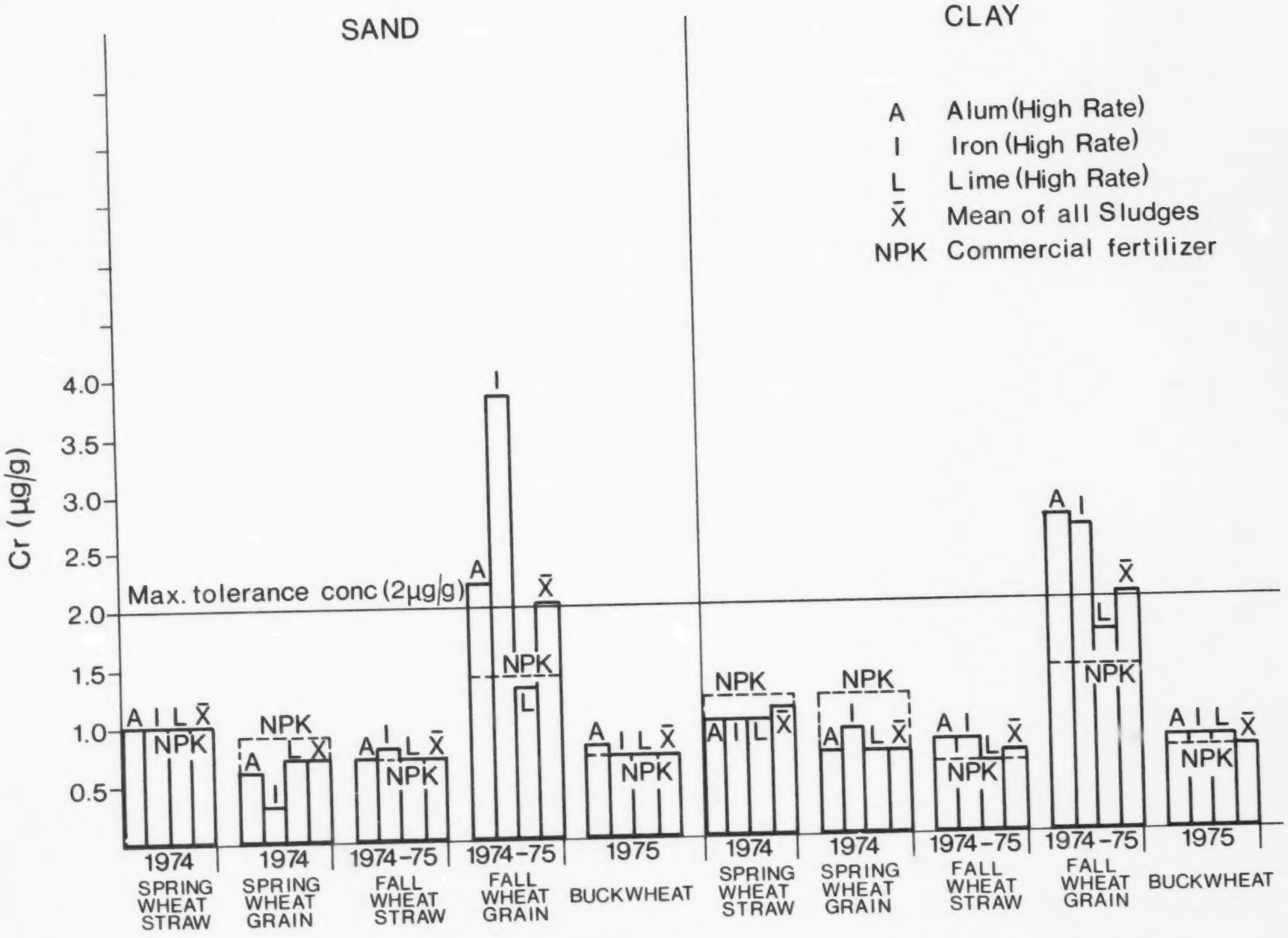


Figure 14 CHROMIUM CONCENTRATION IN PLANT TISSUE (GRAIN/STRAW) (1974 - 75)

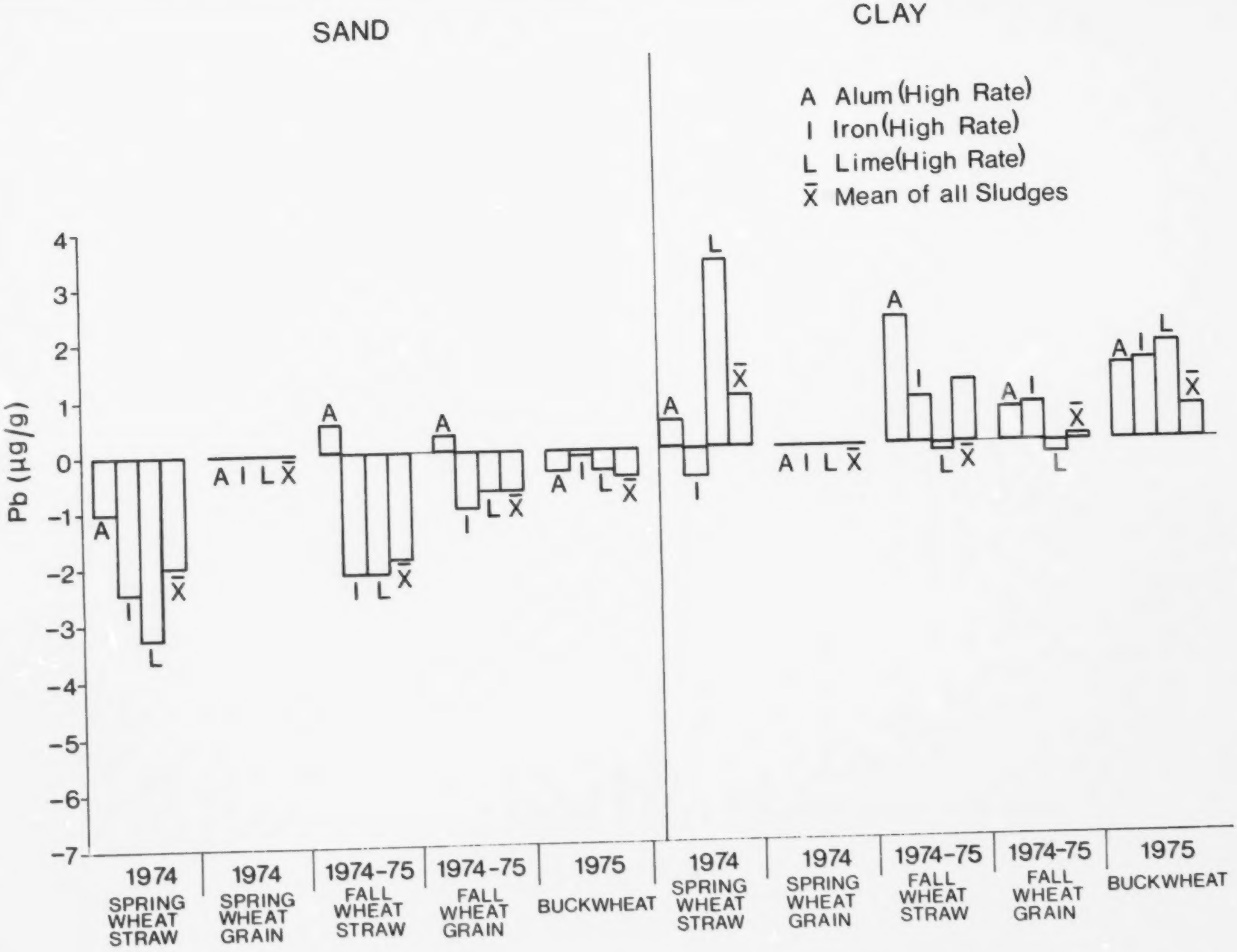


Figure 15 LEAD CONCENTRATION IN PLANT TISSUE (GRAIN/STRAW) (1974 - 75)

(Deviation From NPK Control)

from a major highway adjacent to the lysimeter test site rather than plant uptake. Analysis of forage crops in more rural areas of Ontario and brome grass from the University of Guelph test plots appear to support this hypothesis.

5.3.2.2 Macronutrients. Nitrogen (N), phosphorus (P), and potassium (K) are essential for plant growth and development, and concerns about these elements are related to either:

- deficiencies (limiting crop yields);
- excesses (polluting groundwater);
- germination inhibition (NO_2-N and NH_4-N); or
- toxicity to animals (NO_3-N).

The commercial value of certain crops (e.g., wheat grain) may increase or decrease in direct proportion to the nitrogen (crude protein) content and the intended use of the flour for bread or pastry.

High phosphate concentrations in soils have been associated with the elimination of Zn and Cu toxicities in plants (Spencer, 1966).

Potassium is frequently deficient in soils and sludges, and must be supplemented at high sludge loadings to prevent nitrogen induced potassium deficiencies in crops.

Nitrogen. Nitrogen (N) concentrations in orchard grass grown in lysimeters without NPK fertilizer averaged 2.15% on both soils for all 14 cuts harvested between 1973 and 1976. Addition of sludges (mean of all treatments) increased this concentration to 2.3% on both soils, while NPK fertilizer treatment on both soils averaged 2.55%. The highest nitrogen concentration (2.7%) was obtained with the highest rate of iron sludge applied to loamy sand.

Nitrogen concentration in wheat straw was considerably lower than in orchard grass, averaging only 1.0% for NPK treatment and 1.5% for mean of all sludges on both sand and clay soils. Wheat grain N concentrations were significantly higher than either orchard grass or wheat straw (Figure 16), averaging 3.3% for NPK treatment and 4.0% for mean of all sludge treatments with a maximum of 4.3% for the high rate of alum sludge on sandy soil. Deviations from NPK control showed no

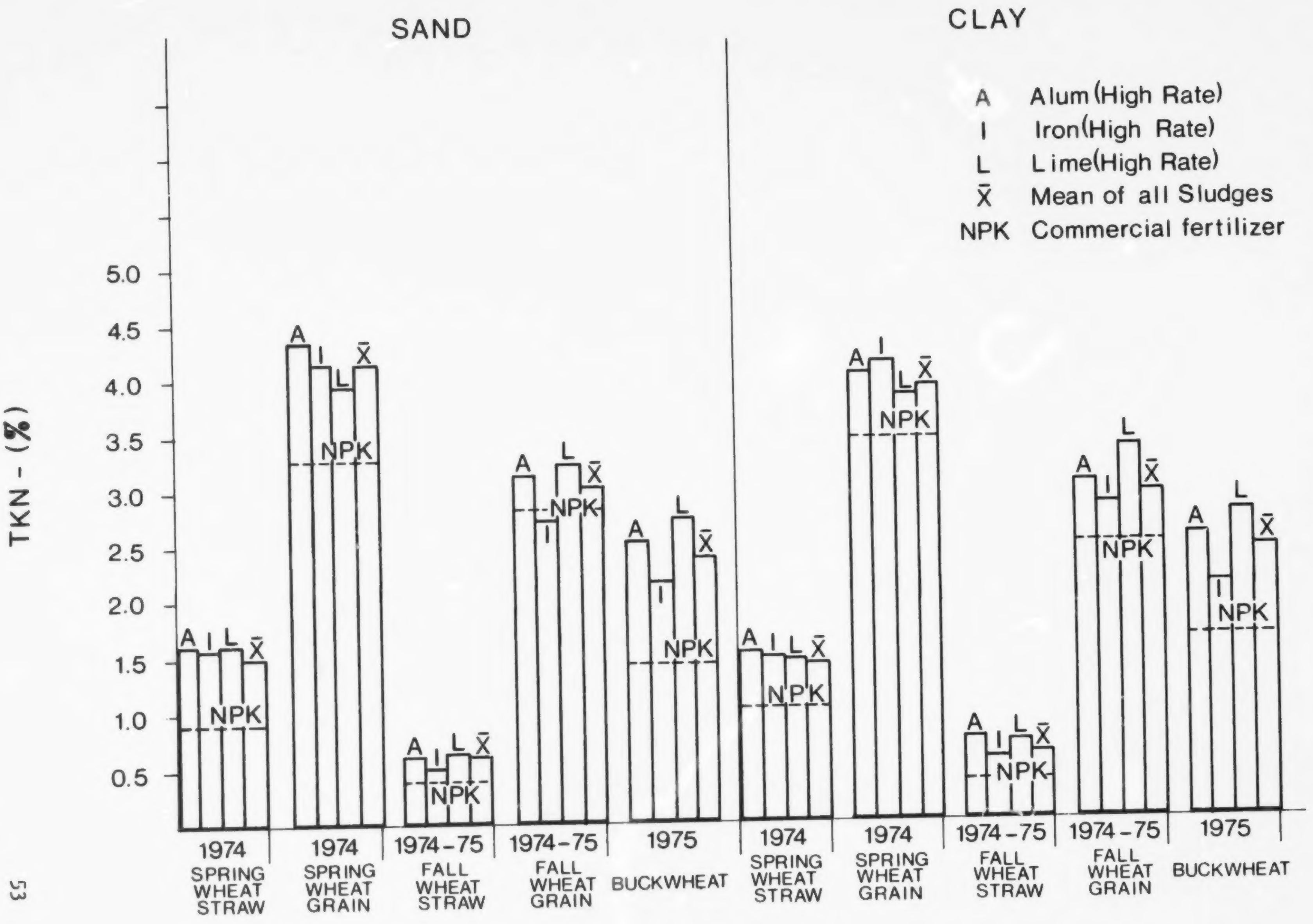


Figure 16 TOTAL NITROGEN CONCENTRATION IN PLANT TISSUE (GRAIN/STRAW) (1974-75)

consistent trend with orchard grass, but were always positive for wheat straw, grain, and buckwheat.

Phosphorus. Phosphorus (P) concentrations in orchard grass (mean of all sludges) increased with time from 0.23% in 1973 to 0.34% in 1976 on loamy sand, with a similar trend observed on silt loam. Unlike heavy metals, P concentrations tended to decrease in plant tissue with increasing sludge loadings. Phosphorus concentrations in spring wheat straw (0.1% for mean of all sludges) were significantly lower than the mean for orchard grass, but P concentrations in wheat grain were higher than in either forage crop, averaging 0.55% on both soils (Figure 17). Deviation from NPK control was always positive for orchard grass ranging from +0.02 to +0.11%. The greatest increase over NPK treatment was observed for buckwheat in sandy soil and averaged +0.30% for all sludge treatments.

Potassium. Potassium (K) concentrations in orchard grass declined from 2.0% on loamy sand in 1973 (mean of all sludge treatments) to only 0.8% in 1974. Concentrations under 1.0% are considered to be seriously deficient. Supplemental applications of 200 kg K/ha in both 1974 to 1975 and 1975 to 1976 raised the (K) mean concentration to 1.4% in 1975 and 1.5% in 1976. A similar trend, although less pronounced, was observed on silt loam.

Deviation from NPK control (Figure 18) increased in proportion to orchard grass plant yield, reaching a maximum of -0.8% in 1974 on loamy sand and -0.9% in 1976 on silt loam (iron sludge - high rate). Deviation from NPK control for wheat straw, grain, and buckwheat on sand and clay soils (Figure 19) differed from orchard grass trends. Virtually no deficiencies were observed on clay soil for any crop. On sandy soil, the straw and buckwheat crops K values declined relative to NPK control but the wheat grain K concentrations were slightly higher than NPK control (+0.05%) for both crops on both soils.

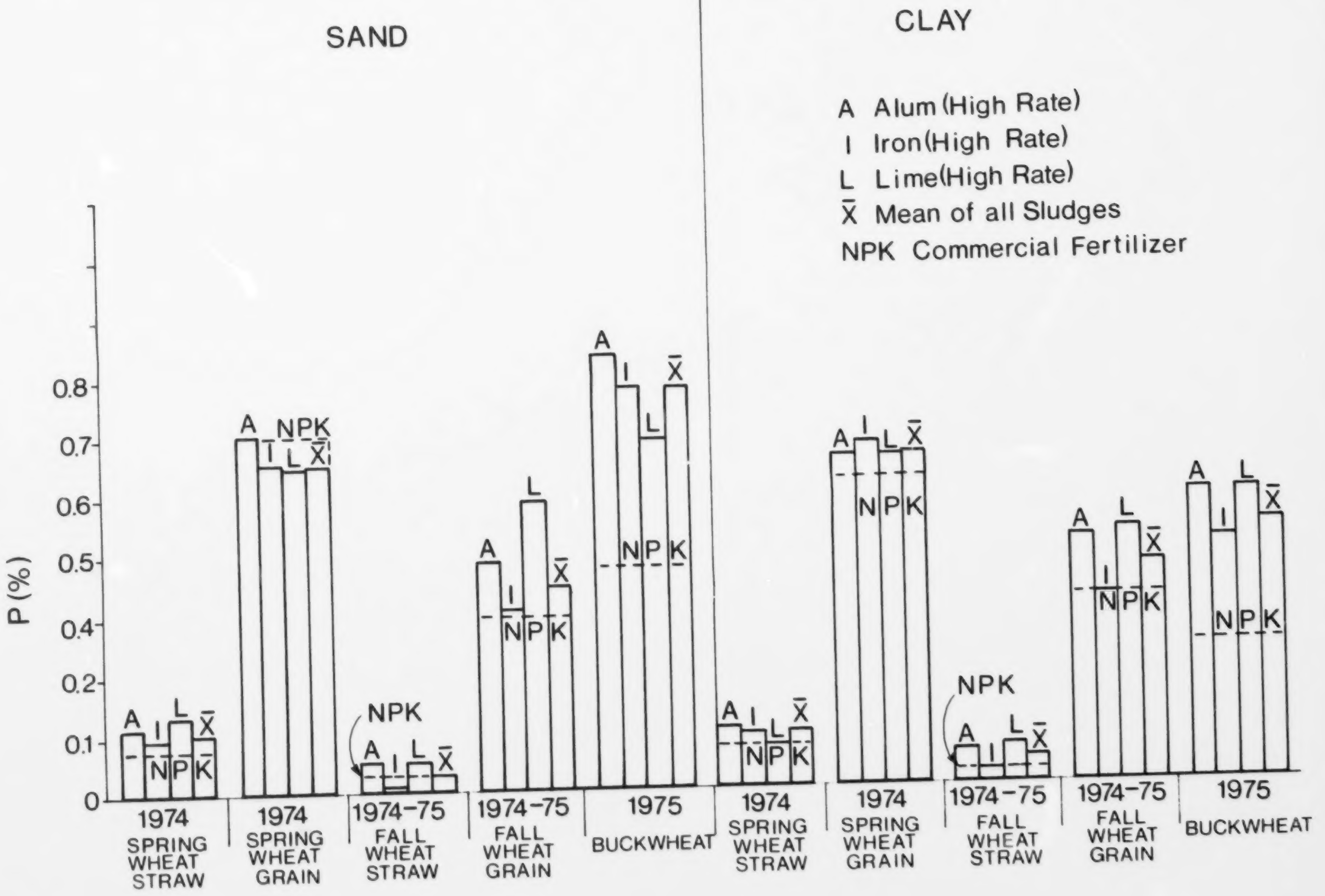


Figure 17 PHOSPHORUS CONCENTRATION IN PLANT TISSUE (GRAIN/STRAW) (1974-75)

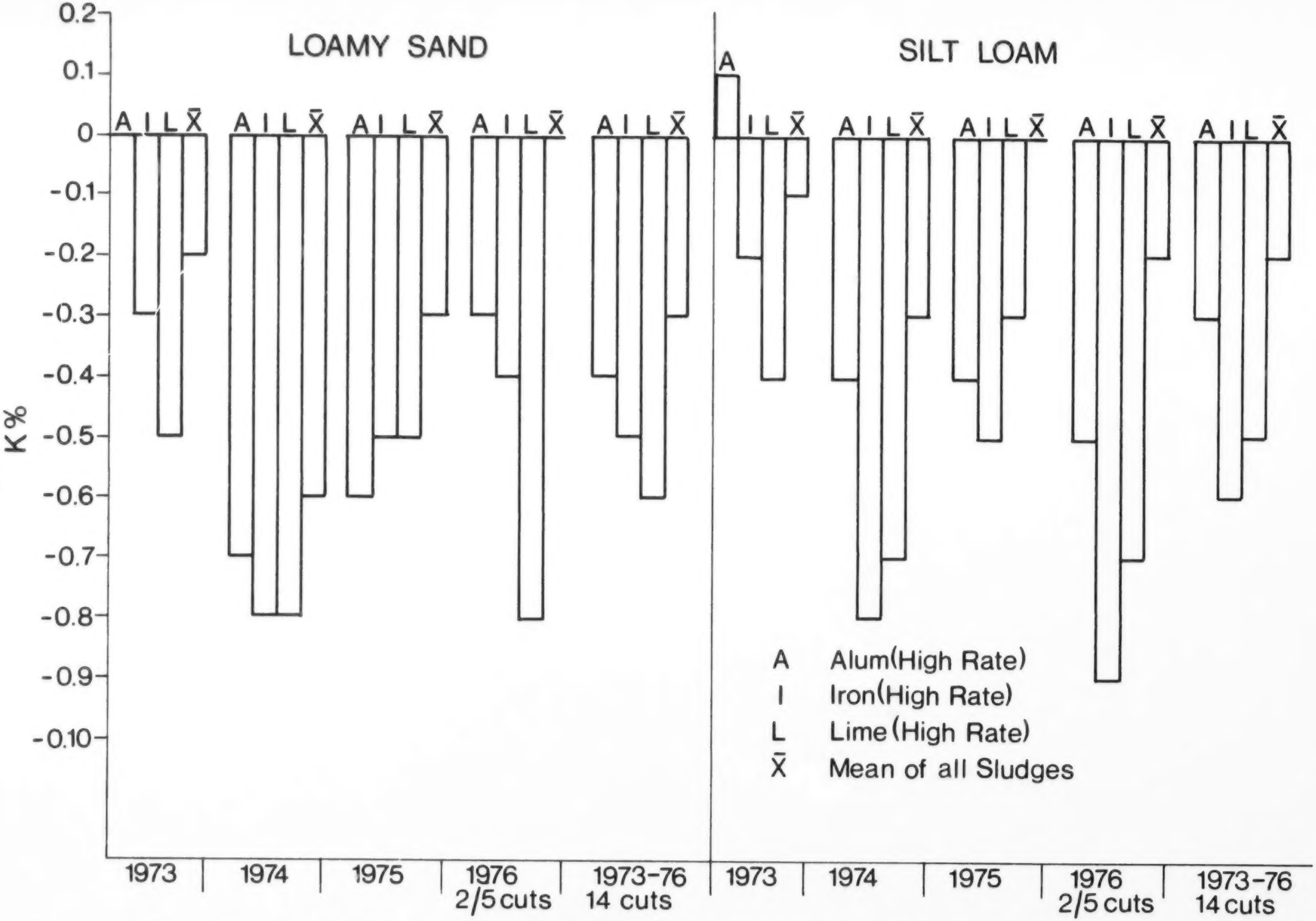


Figure 18 POTASSIUM CONCENTRATION IN ORCHARD GRASS (1973 - 76)

(Deviation From NPK Control)

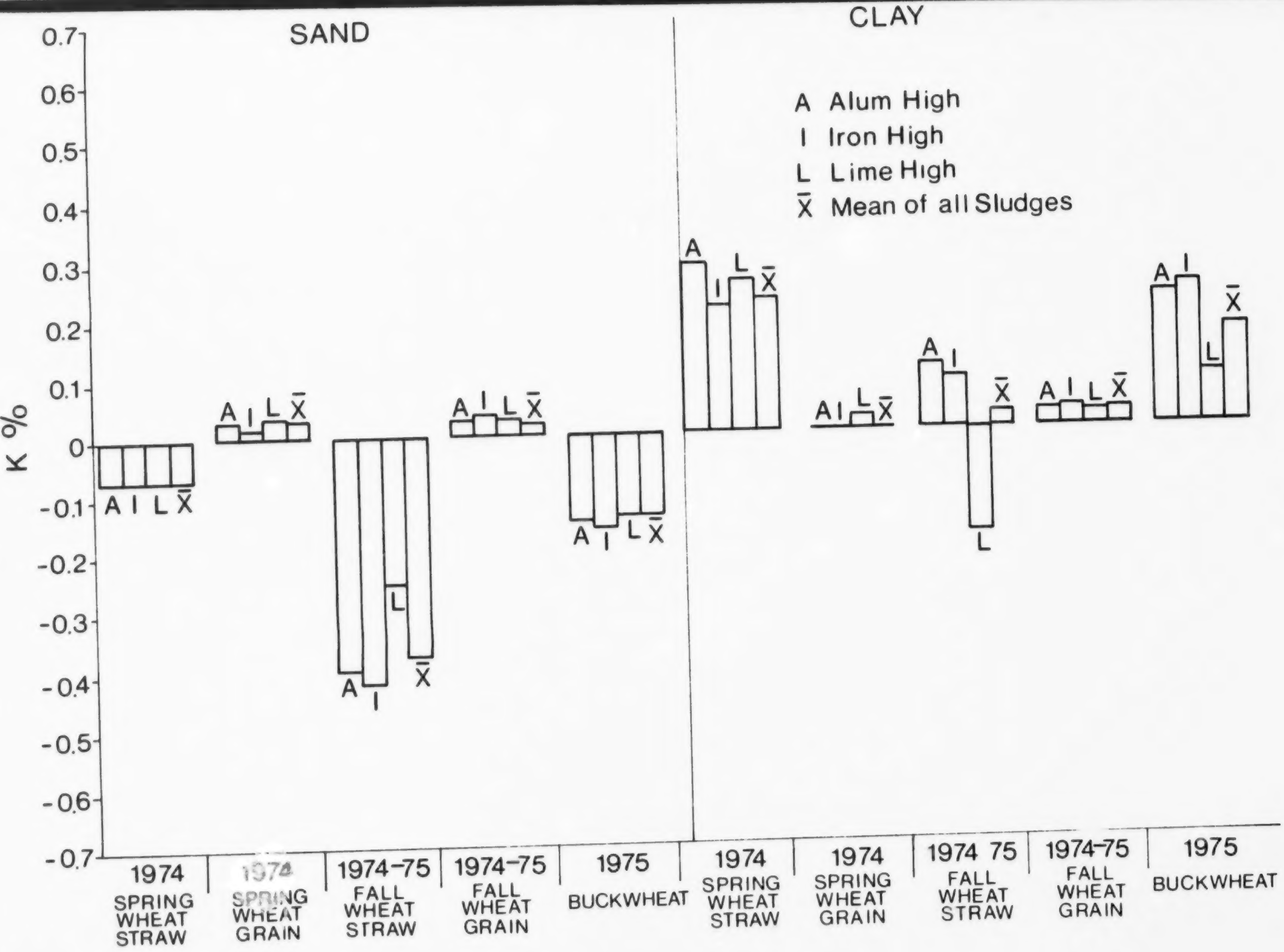


Figure 19 POTASSIUM CONCENTRATION IN PLANT TISSUE (GRAIN/STRAW) (1974-75)

(Deviation From NPK Control)

5.3.2.3 Other chemical constituents. Other plant tissue chemical constituents analyzed included calcium, magnesium, sodium, iron, manganese and aluminum. As with the macronutrients and trace metals, concentrations of these elements can range from deficient through adequate to toxic, depending upon plant species. No significant concentration increases with time were observed for any of the above mentioned constituents.

Concentrations of Fe in orchard grass increased with increasing sludge application rates during a single growing season, but not from one year to the next. Manganese concentrations, in a similar fashion as K, tended to decrease with time and increasing rates of sludge application but remained within the normal range (40 to 150 μg/g) for orchard grass. Aluminum concentrations tended to decline with time for both NPK and sludge treatments but no significant differences were observed due to soil, sludge type or rate. Calcium and magnesium concentrations did not vary significantly with time or sludge rate. Sodium concentrations were in the normal range and were inversely proportional to the potassium concentration deficiencies observed before supplemental K fertilizer was applied.

Air-dried sludge treatments produced wheat grain and straw calcium concentrations that were directly related to application rates. The highest calcium values were obtained on sandy soil. Increased calcium uptake at the highest sludge loadings may have limited heavy metal uptakes, as was previously suggested for phosphorus. Concentrations of Fe, Mn and Al in wheat grain from high rate sludge treatments were comparable to the NPK treatment and were all within acceptable limits for these constituents.

5.3.3 Plant uptake

A well controlled sludge farm will tend to maximize uptake of those macronutrients (NPK) which contribute to high yields and which, if not taken up by plants, can contaminate groundwater. Conversely, plant uptake of "metals of concern" should be limited to avoid plant and animal toxicity problems. The actual quantities of chemical constituents taken up by plants (kg/ha) reflects both concentrations (μg/g) in plant tissue and yields (tons/ha). Both concentrations and yields during a single growing season tended to increase with sludge loading rates. Table 18 shows the maximum orchard grass and wheat uptakes obtained from any sludge treatment at the highest loading rates to all four soils.

TABLE 18. CUMULATIVE PLANT UPTAKE OF CHEMICAL CONSTITUENTS FROM WTC LYSIMETER SLUDGE AMENDED SOILS (kg/ha) MAXIMUM VALUE AT HIGHEST LOADING RATES

Constituent	Fluid Sludge (Orchard Grass) (1973+74+75)		Air-Dried Sludge (Wheat/Buckwheat) (1974+75)	
	Loamy Sand	Silt Loam	Sand	Clay
N	1450	1730	860	1080
P	150	172	102	117
Zn	4.60	4.20	0.84	1.05
Cu	0.59	0.63	0.13	0.25
Ni	0.08	0.10	0.03	0.04
Cr	0.06	0.07	0.03	0.04
Pb	0.60	0.62	0.15	0.19
Cd	0.02	0.02	0.01	0.01

According to the OMAF provisional guidelines (1976), less than 0.3 kg/ha/yr of any of the toxic metals are removed as plant tissue when "normal" concentrations of these elements exist in the plough depth layer of the soil. Of all the metals monitored, only zinc was taken up by orchard grass in amounts that exceed the "norm". Differences in metals uptake between soils growing the same crop was not significant.

Absolute quantities of zinc taken up by the combined wheat/buckwheat harvests of 1974 and 1975 also exceeded 0.3 kg/ha/yr on both soils. The increased uptake of Zn on clay compared to sandy soil reflected the higher yields obtained from clay soil. The only metal whose uptake on clay exceeded the yield differential was copper, but the absolute amounts of copper taken up per year from both soils was low.

Having considered the absolute amounts of chemical constituents taken up by the orchard grass and wheat/buckwheat systems that received sludge over several years, the obvious question is - how do these values differ from normal agronomic uptakes when NPK fertilizers are used (NPK control). Table 19 compares the ratio - maximum uptake at highest loading rate/NPK controls.

TABLE 19. RATIO OF CUMULATIVE PLANT UPTAKE (kg/ha) FROM WTC LYSIMETER SLUDGE AMENDED SOILS (HIGH RATE) COMPARED TO NPK CONTROL

Constituent	Fluid Sludge (Orchard Grass) (1973+74+75)		Air-Dried Sludge (Wheat/Buckwheat) (1974+75)	
	Loamy Sand	Silt Loam	Sand	Clay
N	1.26	1.25	1.51	1.71
P	1.53	1.56	1.57	1.89
Zn	2.00	1.75	1.17	1.75
Cu	2.09	1.66	1.19	1.47
Ni	1.42	1.44	1.15	1.03
Cr	1.50	1.58	1.45	1.38
Pb	1.88	1.38	0.88	1.36
Cd	1.36	1.27	0.64	1.33

At the highest rate of sludge application, approximately twice as much zinc and copper were taken up by orchard grass during 1973 to 1975 than from the NPK control. Increased yields of 38% on loamy sand and 28% on silt loam at the highest iron sludge rate, account for part of this difference. Significantly more zinc and copper were, however, removed per unit of orchard grass harvested from sludge application. Nickel, chromium, and cadmium uptakes, after correction for yield differences did not show (per unit of grass harvested) any significant increase between maximum sludge treatments and NPK control.

Nitrogen uptake was higher, compared to NPK control, on all four soils from the highest sludge loading rate, whereas metal uptake ratios $\left(\frac{\text{sludge-max.rate}}{\text{NPK control}}\right)$ invariably exceeded the nitrogen ratio of 1.25

on both soils growing orchard grass. The metal uptake ratios from the air-dried sludge-wheat experiment except for zinc (1.75), on clay soil, were usually less than the nitrogen uptake ratio. When soluble nitrogen supply limits growth and development, as was the case for air-dried sludge applied to sandy soil, metals uptake is apparently inhibited. Lead and cadmium uptakes at the highest sludge application rate to sandy soil were only 88% and 64%, respectively of the NPK uptakes. For clay soil, however, where crop yields from sludge and NPK were approximately equal, lead and cadmium uptakes were 36% and 33% higher from sludge treatment than from NPK.

To determine whether nitrogen or sludge metal loadings had a greater impact on plant metal uptakes, the ratios of sludge constituent loadings at the highest application rate (kg/ha) were compared with the ratio of plant metal uptakes for the three sludge types, alum, iron and lime. Both sludge and plant uptake ratios were calculated by keeping the alum sludge value equal to 1.0. Table 20 compares the sludge and plant ratios for the fluid sludge-orchard grass system, while Table 21 compares these ratios for the air-dried sludge-wheat system.

In most cases, the uptake of metals from alum, iron and lime sludges reflects plant TKN uptake ratios rather than the sludge metals loading ratios. For example, the ratios of fluid sludge chromium (Cr) loadings were:

Alum : Iron : Lime 1.0 : 3.07 : 0.47
The plant uptake Cr ratios were: 1.0 : 1.09 : 0.82 (L. Sand)
1.0 : 1.11 : 0.86 (S. Loam)
The plant uptake TKN ratios were: 1.0 : 1.29 : 0.97 (L. Sand)
1.0 : 1.14 : 0.95 (S. Loam)

Similarly, the ratio of air-dried sludge chromium loadings were:

Alum : Iron : Lime 1.0 : 2.16 : 0.74
The plant uptake Cr ratios were: 1.0 : 1.28 : 0.76 (Sand)
1.0 : 0.89 : 0.69 (Clay)
The plant uptake TKN ratios were: 1.0 : 0.81 : 1.04 (Sand)
1.0 : 0.80 : 0.92 (Clay)

Figure 20 depicts these plant uptake ratios for lead and TKN in orchard grass compared to sludge loading ratios.

TABLE 20. RATIO OF CUMULATIVE TKN AND HEAVY METAL PLANT UPTAKES FROM FLUID SLUDGE AMENDED SOILS COMPARED TO LOADING RATIOS OF ALUM, IRON AND LIME SLUDGES (HIGH RATE)

Constituent	Sludge Loading Ratio			Plant Uptake Ratio					
				Loamy Sand			Silt Loam		
	Alum[1]	Iron	Lime	Alum	Iron	Lime	Alum	Iron	Lime
Zn	1.0	2.32	1.70	1.0	1.34	0.95	1.0	1.21	0.87
Cu	1.0	1.73	0.76	1.0	1.21	0.88	1.0	1.17	0.85
Ni	1.0	0.88	1.25	1.0	1.24	0.92	1.0	1.35	0.97
Cr	1.0	3.07	0.47	1.0	1.09	0.82	1.0	1.11	0.86
Pb	1.0	5.40	1.16	1.0	1.43	0.98	1.0	1.27	0.96
Cd	1.0	2.63	1.63	1.0	1.25	1.13	1.0	1.10	0.77
Mean of 6 Metals ($\bar{x}$)	1.0	2.67	1.16	1.0	1.26	0.95	1.0	1.20	0.88
TKN	1.0	1.0	1.0	1.0	1.29	0.97	1.0	1.14	0.95

[1] Alum sludge (kg/ha = 1.0).

TABLE 21. RATIO OF CUMULATIVE TKN AND HEAVY METAL PLANT UPTAKES FROM AIR-DRIED SLUDGE AMENDED SOILS COMPARED TO LOADING RATIOS OF ALUM, IRON AND LIME SLUDGES (HIGH RATE)

Constituent	Sludge Loading Ratio			Plant Uptake Ratio					
				Sand			Clay		
	Alum[1]	Iron	Lime	Alum	Iron	Lime	Alum	Iron	Lime
Zn	1.0	1.06	1.13	1.0	1.0	0.94	1.0	0.96	0.80
Cu	1.0	1.05	0.89	1.0	0.91	1.0	1.0	0.69	0.71
Ni	1.0	0.35	0.97	1.0	0.93	0.74	1.0	1.23	1.37
Cr	1.0	2.16	0.74	1.0	1.28	0.76	1.0	0.89	0.69
Pb	1.0	3.25	1.30	1.0	0.78	0.89	1.0	0.92	0.94
Cd	1.0	1.53	1.0	1.0	0.86	1.0	1.0	1.33	1.0
Mean of 6 Metals ($\bar{x}$)	1.0	1.57	1.0	1.0	0.96	0.89	1.0	1.0	0.92
TKN	1.0	0.70	1.94	1.0	0.81	1.04	1.0	0.80	0.92

[1] Alum sludge (kg/ha = 1.0).

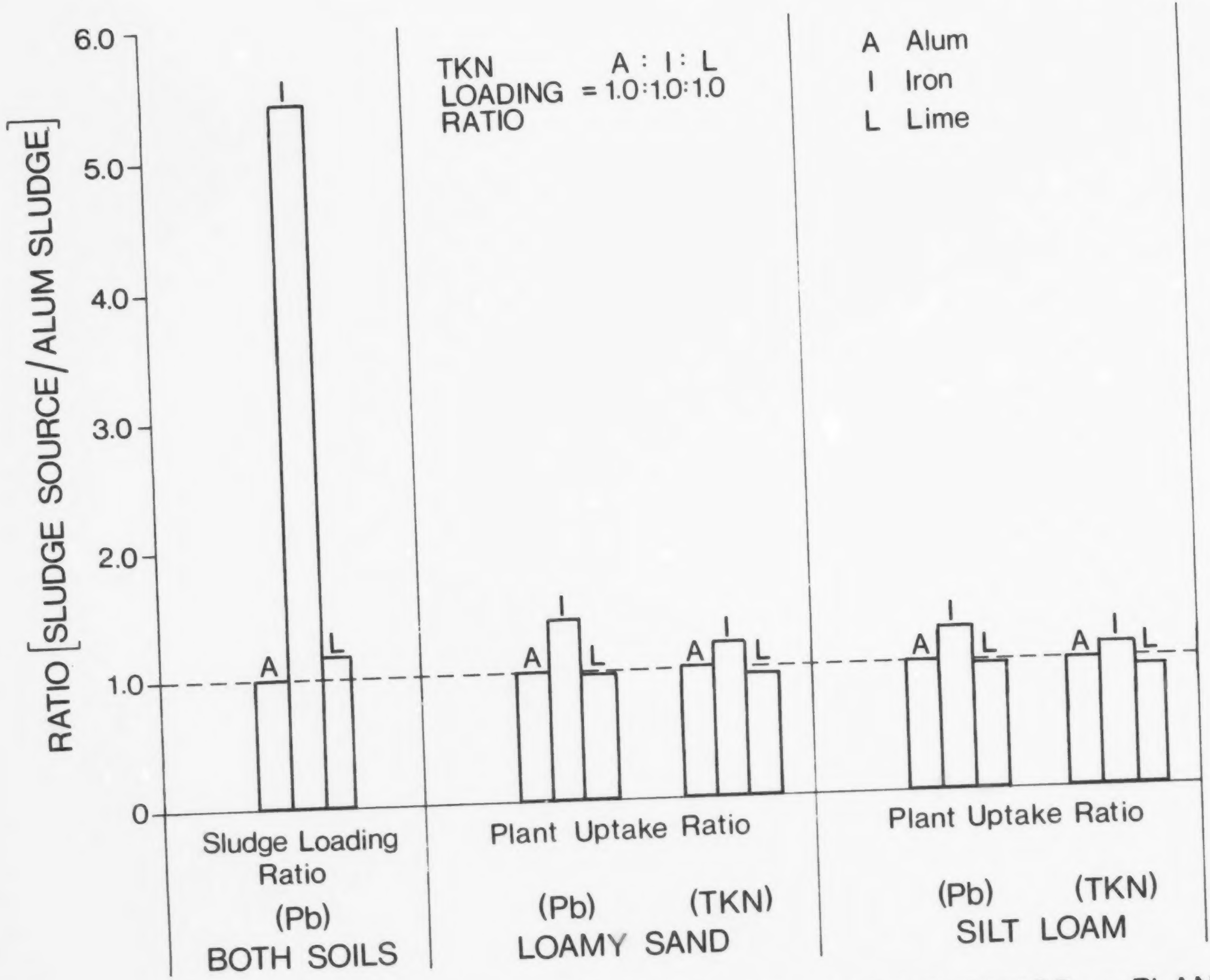


Figure 20 LEAD AND TKN FLUID SLUDGE LOADING RATIOS vs PLANT (ORCHARD GRASS) UPTAKE RATIOS

The OMAF provisional guidelines (1976) for application of fluid sludge to farmland relate acceptable metal concentrations to plant-available nitrogen content. Thus, a minimum ratio of NH_4-N/metal must be maintained if the sludge is to be accepted as suitable for agricultural use. Table 22 compares suitability of the alum, iron, and lime, fluid and air-dried sludges used in the lysimeter experiments with this guideline. With minor exceptions, the fluid sludges would be accepted and the air-dried sludges rejected. It may be inferred from this guideline that the nitrogen/metal ratios of the fluid sludges provide a larger safety factor against unacceptable metals uptake by plants than do the air-dried sludge ratios. One might expect to see this difference reflected in the nitrogen/metal ratios of the plants grown on soils receiving either fluid or air-dried sludge. Table 23 compares the nitrogen/metal ratios of the orchard grass crop (fluid sludge) and wheat/buckwheat total crop (air-dried sludge) based on cumulative plant uptakes (kg/ha) at the highest loading rates. As soil differences did not significantly influence the N/metal uptake ratios, a single average value for each crop is given.

TABLE 22. ASSESSMENT OF WTC SLUDGE SUITABILITY FOR AGRICULTURAL LAND APPLICATION

Constituent	Minimum Ratio[1] Required	Fluid Sludge (1973+74)			Air-Dried Sludge (1974)		
		Alum	Iron	Lime	Alum	Iron	Lime
Zn	4	8.35	4.5	4.9	0.85	0.25	0.42
Cu	10	12.3	9.4	16.2	1.7	0.5	1.0
Ni	40	278	412	213	88	30.4	18.5
Cr	15	48	21	106	5.3	0.9	3.6
Pb	15	29	8	27	3.2	0.3	1.1
Cd	500	835	515	567	74	21	27
Acceptability	-	6 out of 6	4 out of 6	6 out of 6	1 out of 6	0 out of 6	0 out of 6

[1] Ratio NH_4-N/Metals; Ref: OMAF, 1976.

TABLE 23. NITROGEN/METAL RATIOS OF ORCHARD GRASS AND WHEAT/BUCKWHEAT CROPS BASED ON CUMULATIVE PLANT UPTAKE (kg/ha) AT THE HIGHEST SLUDGE LOADING RATE[1]

Constituent	Fluid Sludge (Orchard Grass) (1973+74+75)	Air-Dried Sludge (Wheat/Buckwheat) (1974+75)	Ratio $\frac{\text{Air-Dried Sludge}}{\text{Fluid Sludge}}$
Zn	364	1,028	2.82
Cu	2,602	5,468	2.10
Ni	17,713	27,834	1.57
Cr	24,440	27,834	1.14
Pb	2,604	5,709	2.19
Cd	79,500	97,000	1.22

[1] N/Metal ratio derived from mean of two soils (Table 18).

Different plant species were grown on the fluid and air-dried sludge amended soils. The heavy metal concentrations in orchard grass (Table 14) wheat straw (Table 16) and buckwheat (Table 17) were similar enough to discount this factor as explaining the higher N/metal ratios observed for all six metals in plant tissue from the air-dried sludge lysimeter experiment.

From the above discussion, the NH_4-N/metal ratio criteria presently proposed for fluid sludges may not be appropriate for air-dried sludges. The justified attempt to limit plant heavy metals uptake by relating soluble nitrogen needs of the crop to sludge metal concentrations, would, therefore, be unduly restrictive unless allowance is made for a reasonable, experimentally verified, organic nitrogen mineralization rate for dry sludges.

5.3.4 Plant recovery efficiency

Plant recovery efficiency is defined as that fraction of a chemical constituent contained in sludge applied to soil that is recovered in the total plant tissue harvested. Table 24 summarizes the recovery efficiency for the elements N, P, Zn, Cu, Ni, Cr, Pb, and Cd, based on sludge constituent loadings at the highest rate, and cumulative plant uptake maximum values.

TABLE 24. PLANT RECOVERY EFFICIENCY (%) OF NUTRIENTS AND HEAVY METALS AT MAXIMUM SLUDGE LOADING RATES

Constituent	Fluid Sludge (Orchard Grass) (1973+74+75)		Air-Dried Sludge (Wheat/Buckwheat) (1974+75)	
	Loamy Sand	Silt Loam	Sand	Clay
Total N*	40.2	48.1	12.3 - 34.6	15.4 - 43.4
Total P	4.46 - 8.14	5.11 - 9.33	1.27 - 3.32	1.45 - 3.81
Zn	2.17 - 5.05	1.98 - 4.62	0.48 - 0.54	0.60 - 0.68
Cu	0.58 - 1.30	0.62 - 1.40	0.15 - 0.18	0.30 - 0.35
Ni	2.60 - 3.71	3.27 - 4.67	0.88 - 2.50	1.21 - 3.42
Cr	0.13 - 0.85	0.15 - 1.01	0.08 - 0.23	0.09 - 0.26
Pb	0.44 - 2.40	0.46 - 2.48	0.13 - 0.42	0.16 - 0.53
Cd	0.71 - 1.88	0.90 - 2.38	0.30 - 0.47	0.34 - 0.53

* Mean values.

Orchard grass recovery efficiency maximum values were higher from soil with the higher cation exchange capacity (silt loam) for all constituents except zinc. Similarly, wheat/buckwheat recovery efficiencies were always greater on clay than on sandy soil. Recovery efficiencies greater than 2% were observed for zinc, nickel, lead and cadmium from orchard grass, but only nickel recovery exceeded 2% for the wheat/buckwheat system.

A comparison of mean recovery efficiencies and dry matter yields (two soils x three sludge types) of orchard grass and wheat/buckwheat at the maximum sludge loading rate is presented in Table 25.

Total recovery of all constituents was significantly greater in orchard grass than wheat/buckwheat. After correcting for the 133% greater yield of orchard grass than buckwheat, and calculating recovery on a unit weight basis, significantly more nitrogen and nickel were recovered from the wheat/buckwheat crop than from orchard grass. Zinc total recovery was nearly six times greater in orchard grass than in wheat. Copper, cadmium, and lead recoveries were approximately four times greater in orchard grass. A great deal more information is required concerning cumulative uptake of metals from a wide variety of agricultural crops, receiving sludge in either fluid or dry form. More information concerning the tendency of a particular crop to remove a particular "constituent of concern" from sludge amended soil with a high recovery efficiency could allow for increasing refinement in sludge application guidelines as currently proposed.

TABLE 25. COMPARISON OF ORCHARD GRASS AND WHEAT/BUCKWHEAT RECOVERY EFFICIENCIES AT MAXIMUM SLUDGE LOADING RATE (MEAN OF 2 SOILS X 3 SLUDGE TYPES) vs PLANT DRY MATTER YIELDS

Constituent	Mean Recovery Efficiency (%)		
	Orchard Grass (1973-75)	Wheat/Buckwheat (1974-75)	Ratio Orchard Grass / Wheat/Buckwheat
N	44.2	26.4	1.67
P	6.76	2.46	2.75
Zn	3.46	0.58	5.97
Cu	0.98	0.25	3.92
Ni	3.57	2.00	1.79
Cr	0.54	0.17	3.18
Pb	1.45	0.31	4.67
Cd	1.47	0.41	3.59
Plant Dry Matter Yield Cumulative - tons/ha	70.5	30.2	2.33

5.4 Leachate

For each intended use of water (drinking, irrigation, etc.) there exist well defined and generally accepted quality criteria. Criteria for chemical constituents of concern in soils, sludges and plants, on the other hand, are less clearly defined.

Requirements for sewage effluents and land drainage in Ontario (OMOE, 1974) are based on water quality standards or criteria to maintain acceptable water quality. Because of limitations in water management information, a margin of safety is provided in water quality standards.

The responsibility for demonstrating that a waste effluent (such as leachate from sludge amended soil) is harmless to water uses, in the concentrations found in the receiving waters, rests with those producing the discharge.

For groundwater quality surveillance, unlike that for surface waters supporting aquatic life, the average concentration of a chemical constituent over a considerable period of time is significant rather than a limit which may not be exceeded at any time.

There are distinct differences between short and long term consequences in the impact of sludge application to agricultural soils from the standpoint of irrigation water quality. Maximizing crop yields requires preventing the buildup of excessive amounts of plant available salts or specific elements in the soil. Thus, in the short run, leaching excess soluble constituents of concern from the soil is desirable. In the long run, the need to use the groundwater for irrigation requires protecting it against contamination from sludged soil leachate constituents, to ensure that maximum permissible concentrations will not be exceeded. Thus groundwater protection may require a minimum of leaching.

Table 26 summarizes water quality criteria for various agricultural, industrial and public (potable) uses in Ontario (OMOE, 1974). It should be borne in mind that these maximum permissible concentrations cannot be considered as maximum single sample values. The guidelines require that, if these criteria are exceeded "over substantial portions of time", the cause must be determined and corrective action initiated. From this table, drinking water criteria may be seen to be most restrictive (with the exception of those for copper for dairy or textile use). Pathogenic, radioactive, or pesticide criteria are not included as they are beyond the scope of this paper.

Very few studies that quantify the quality of leachate from sludge amended soils are available. In one study quoted by Bates (1972), the Greater Chicago Sanitary District (GCSD) monitored groundwater for four years after application of 340 dry tons/ha of digested sludge on two different sites. No increase in nitrate levels over normal background concentrations were detected in test wells, although an increase of up to 7 mg NH_4-N/l was observed in wells on one site.

TABLE 26. WATER QUALITY CRITERIA FOR VARIOUS USES (OMOE, 1974) MAXIMUM PERMISSIBLE CONCENTRATIONS (mg/l)

Constituent or Characteristic	Agriculture	Industry	Public (Drinking)
I Metals			
Cu	0.1(d) ; 5.0(i)	0.01(t)	1.0
Mn	; 20.0(i)	0.05(b)	0.05
As	0.05(l) ; 10.0(i)		0.05
Cd	0.01(l) ; 0.05(i)		0.01
$Cr^{(6+)}$	0.05(l) ; 20.0(i)		0.05
Pb	0.05(l) ; 20.0(i)		0.05
Al	; 20.0(i)		-
Ni	; 2.0(i)		-
Zn	; 5.0(i)		5.0
II Plant Macronutrients			
K	20(d)		
NO_2-N + NO_3-N	20.0(i)	10.0(c)	10.0
NH_4-N			0.5
P			0.1
III Other Characteristics			
Organic Matter (Carbon chloroform extract)		0.15(b)	0.15

(d) dairy; (l) livestock; (i) irrigation; (b) brewing; (c) chemicals; (t) textile.

Ammonium levels at this site subsequently declined without any discernible nitrification effect being noted. Hinesly and Sosewitz (1969), commenting on the work done by the GCSD, state that sludge application rates which supply nitrogen in amounts greater than those needed for crop production will increase nitrate levels in drainage water. From the information mentioned above, one cannot easily predict where, when, whether or to what extent "increased nitrate levels in drainage water" will increase nitrate levels in groundwaters receiving these drainage waters.

Phosphorus (P) in sludge incorporated in soils has not usually been a water pollution hazard (Sopper, 1971). Most agricultural soils have a tremendous capacity to hold phosphorus in insoluble forms, although Huseman and Wesche (1962) provide some evidence that phosphorus groundwater pollution might occur at very high rates of sludge application to either coarse (sandy) or organic soils.

The heavy metals Cu, Cr, Cd, Ni, Pb and Zn are all held in water insoluble forms by soils. There is some evidence (Kick et al, 1963) that the organic matter (particularly chelating compounds) in sludges hold these metals in very stable complexes, thus correcting toxic effects of heavy metals in soils from non-sludge sources. The heavy metals - chelate complexes are generally more soluble and available for plant uptake or leaching when soil pH declines below 6.5. Anaerobic conditions, which can occur when heavy loadings of rapidly decomposing organic matter cause a very high oxygen demand in the soil, may also increase the solubility of heavy metals. Thus, if the heavy metals monitored in these experiments are to be prevented from contaminating groundwater, a well aerated neutral to alkaline soil is required. This requirement could conflict with efforts to prevent nitrate pollution of groundwater from sludged soil by flooding to enhance denitrification.

Although trace elements are relatively immobile in soil, nickel and zinc have been shown to move below the 30 cm depth, while chromium and mercury have been reported to move below 120 cm (Kirkham, 1976). Where water tables are high, plants on sludge treated land might conceivably take up trace elements from contaminated groundwater. The time required for a particular contaminant to leach into underlying groundwater may be decades but, once the damage is done, correction is extremely difficult. A case in point is the incident in Minnesota in 1972 (Josephson, 1976) in which arsenic contamination of well water occurred, 38 years after an arsenic based pesticide had been massively applied to deal with a grasshopper infestation.

The little information that is available concerning concentration of trace elements in drainage water from sludge amended sites is inconclusive or even contradictory. In a Chicago study of leachate from corn plots at Hanover Park, Illinois, up to 203 tons/ha dry solids were cumulatively applied over six years. All nutrients and trace metals monitored declined in concentration with time (Lynam, 1974). A Canadian study of well water quality from sludge amended land near Toronto (Van Loon, 1975) showed large variations in iron and zinc values, but little variation in copper and nickel concentrations. A British study (Pike et al, 1975), at a long term sludge disposal site, showed nickel to be far more mobile than the other heavy metals (Cd, Cu, Cr, Pb, Zn) monitored. In a German study, El-Bassam et al (1975) found no evidence of either chromium or mercury contamination of groundwater after 80 consecutive years of sewage irrigation and sludge farming. Obviously, more information is needed concerning the extent and conditions under which trace elements migrate in soil of varying characteristics.

5.4.1 Leachate volume losses

Water (irrigation plus precipitation) applied in the orchard grass experiment averaged 1500 m^3/ha/month (150 mm) during the period 1973 to 1976. Water application rate during the summer (growing season) was approximately double (2000 m^3/ha/month) the winter (dormant season) rate. During the growing season, volumes leached varied inversely with crop yields and sludge application rates. Leachate recovery as a percentage of total water applied ranged from a high of 35% for the non-fertilized control on loamy sand to a low of only 6% at the high iron sludge application rate to silt loam. Leachate losses from loamy sand were nearly double cumulative losses from the heavier textured silt loam soil. Leachate volume losses from the various sludge treatments exceeded the NPK control losses with the exception of lime sludge at the highest rate on both soils.

Leachate losses from the air-dried sludge-wheat experiment did not vary significantly with sludge loading rate thus reflecting similar crop yields for all fertility treatments on both soils.

Table 27 summarizes cumulative leachate volume losses from all four soils for the period ending in September, 1976.

Table 28 illustrates the inverse relationship between leachate volume loss and crop yield by comparing the ratio of volumes lost at the highest and lowest rates of sludge application.

TABLE 27. CUMULATIVE LEACHATE VOLUME LOSSES ($m^3 \times 10^3$/ha) FROM FLUID AND AIR-DRIED SLUDGE TREATED SOILS

Treatment/Rate	Fluid Sludge (Orchard Grass) (1973-76)		Air-Dried Sludge (Wheat/Buckwheat) (1974-76)	
	Loamy Sand	Silt Loam	Sand	Clay
Alum (low)	16.0	9.2	6.9	5.6
(medium)	11.0	5.0	7.0	7.4
(high)	10.0	5.4	6.9	7.4
Iron (low)	12.5	7.0	7.0	6.6
(medium)	9.6	4.5	6.8	6.4
(high)	7.8	4.3	7.3	6.5
Lime (low)	13.0	7.8	6.8	6.6
(medium)	8.0	3.8	6.2	5.5
(high)	6.2	3.6	5.5	5.0
NPK fertilizer	7.0	3.8	6.8	6.5

TABLE 28. COMPARISON OF LEACHATE VOLUME LOSS RATIO (HIGH RATE/LOW RATE)[1] vs CROP YIELD RATIO (LOW RATE/HIGH RATE)

Sludge	Leachate Loss Ratio[2]				Crop Yield Ratio[3]			
	Highest Rate/Lowest Rate				Highest Rate/Lowest Rate			
Treatment	Loamy Sand	Silt Loam	Sand	Clay	Loamy Sand	Silt Loam	Sand	Clay
Alum	0.63	0.59	1.00	1.32	1.92	1.59	1.09	1.05
Iron	0.62	0.61	1.04	0.98	1.92	1.75	1.10	1.06
Lime	0.48	0.46	0.81	0.76	1.85	1.64	1.41	0.97

[1] Sludge application rate ratio.
[2] Calculated from Table 27 data.
[3] Calculated from Table 11 data.

5.4.2 Leachate chemical constituents

Eighteen chemical constituents have been regularly monitored in all leachate samples collected since 1973. Of these, six heavy metals and four nutrient forms (NO_3-N, NH_4-N, total P and total organic carbon (TOC)) have been compared to existing water quality standards to determine whether leachate quality long term trends indicate any cause for concern. Tables 29 and 30 summarize leachate quality mean values (1973 to 1976) for the fluid sludge-orchard grass and air-dried sludge-wheat experiments (1974 to 1976), respectively, compared to water quality standards.

Leachate heavy metal concentrations (mean of all sludge treatments) from both the air-dried and fluid sludge lysimeters are not significantly different from values observed for NPK treatment on all soils. Maximum values observed from sludge treatments for the following metals are expressed as a percentage of the maximum permissible concentrations (MPC) for drinking water (NH&W, 1969) or irrigation water (OMOE, 1974):

	Max. Ave. Annual Value % of MPC	
	Fluid	Air-Dried
Zinc	0.9	0.8
Copper	7.0	4.2
Nickel	1.7	0.6
Chromium	14.0	54.0
Lead	82.0	58.0
Cadmium	40.0	40.0

The values shown in Tables 29 and 30 are maximum annual means. Within a particular year individual leachate samples from sludge amended soils had concentrations which approached but did not exceed the MPC for drinking water. On occasion, individual samples of leachate had concentrations of Cr, Pb, and Cd from sludge amended soils which exceeded those from non-sludge controls. Maximum values were usually observed

TABLE 29. LEACHATE QUALITY FROM FLUID SLUDGE AMENDED SOILS AND CONTROLS (1973 to 1976) COMPARED TO WATER QUALITY STANDARDS (mg/l)

Constituent	Control				Sludge Amended Soils				Water Quality Standards			
	Non Fertilized		NPK Fertilizer		Mean of All Treatments		Maximum Avg. Annual Value		Drinking Water (a)	Irrigation (b)	Effluent	
	Loamy Sand	Silt Loam	Loamy Sand	Silt Loam	Loamy Sand	Silt Loam	Loamy Sand	Silt Loam			(c)	(d)
NH_4-N	0.08	0.05	0.04	0.04	0.21	0.13	0.60	0.49	0.5			
NO_3-N	4.5	1.2	8.7	3.6	7.0	3.5	19.5	18.5	10.0			
Total P	0.16	0.11	0.07	0.15	0.42	0.19	2.3	0.8	<0.1		1.0	
TOC	7.8	8.2	12.5	18.3	13.3	14.8	19.3	18.1	-			15.0(BOD)
Zn	0.010	0.015	0.020	0.046	0.021	0.034	0.028	0.046	5.0			
Cu	0.011	0.018	0.015	0.043	0.022	0.041	0.031	0.070	1.0	5.0		
Ni	0.005	0.009	0.006	0.010	0.009	0.015	0.019	0.034	-	2.0		
Cr	0.004	0.005	0.003	0.005	0.004	0.006	0.006	0.007	0.05			
Pb	0.013	0.014	0.012	0.020	0.013	0.022	0.021	0.041	0.05	20.0		
Cd	0.002	0.003	0.002	0.003	0.002	0.003	0.003	0.004	0.01			

(a) Source: NH & W (1969), Canadian Drinking Water Standards.
(b) Source: OMOE (1974), Water Quality Criteria for Agricultural Uses.
(c) Source: Canada/US (1972), GLWQ Agreement - Effluent Requirement.
(d) Source: OWRC (1970).

TABLE 30. LEACHATE QUALITY FROM AIR-DRIED SLUDGE AMENDED SOILS AND CONTROLS (1974 to 1976) COMPARED TO WATER QUALITY STANDARDS (mg/l)

Constituent	Control				Sludge Amended Soils				Water Quality Standards			
	Non Fertilized		NPK Fertilizer		Mean of All Treatments		Maximum Avg. Annual Value		Drinking Water (a)	Irrigation (b)	Effluent	
	Sand	Clay	Sand	Clay	Sand	Clay	Sand	Clay			(c)	(d)
NH_4-N	0.14	0.12	0.14	0.10	0.23	0.12	0.40	0.18	0.5			
NO_3-N	15.6	34.8	19.2	41.9	75.0	81.3	157.6	156.8	10.0			
Total P	0.23	0.23	0.21	0.27	0.24	0.18	0.34	0.27	0.1		1.0	
TOC	19.8	55.6	18.8	54.7	29.5	53.8	35.5	57.1	-			15.0(BOD)
Zn	0.042	0.068	0.038	0.021	0.035	0.023	0.042	0.027	5.0			
Cu	0.037	0.021	0.045	0.021	0.034	0.026	0.042	0.027	1.0	5.0		
Ni	0.009	0.005	0.010	0.006	0.009	0.005	0.011	0.006	-	2.0		
Cr	0.010	0.008	0.008	0.012	0.016	0.010	0.027	0.010	0.05			
Pb	0.027	0.012	0.046	0.011	0.022	0.013	0.029	0.013	0.05	20.0		
Cd	0.002	0.002	0.002	0.002	0.003	0.002	0.004	0.002	0.01			

(a) Source: NH & W (1969), Canadian Drinking Water Standards.
(b) Source: OMOE (1974), Water Quality Criteria for Agricultural Uses.
(c) Source: Canada/US (1972), GLWQ Agreement - Effluent Requirement.
(d) Source: OWRC (1970.

during the first year of leachate collection when the soil-plant systems had not yet stabilized. No consistent increase over time has been observed for any heavy metal monitored. Leakage of nutrients from the soil root zone to groundwater have exceeded water quality standards for NO_3-N, NH_4-N and Total P. Although no water quality standard exists for TOC, the equivalent BOD requirement for a well stabilized secondary sewage effluent (15.0 mg/l) has been used to define the desired leachate quality for organic carbon.

Figures 21 and 22 illustrate changes in NO_3-N concentrations with time, in leachate from fluid and air-dried sludge treated soils, respectively.

The maximum permissible NO_3-N concentration (10.0 mg/l) was exceeded in leachate from fluid sludge-orchard grass lysimeters (mean of all sludge treatments) from both soils during the first year of the experiment (Figure 21). Nitrate leakage was minimal during 1974 and 1975. During 1976, an increase in NO_3-N concentration was discernible, particularly for the highest loading rates of alum and iron sludges on both soils. Although the mean NO_3-N value for all sludge treatments on loamy sand (7.0 mg/l) was double the value from silt loam (3.5 mg/l), maximum average annual values were similar for both soils. Lime sludge treatment exhibited lower NO_3-N leachate concentrations than did alum or iron sludges at equivalent loading rates.

The heavy single dose of air-dried sludge applied to sand and clay soils in 1974 exceeded both the organic nitrogen denitrification capacity of these soils and the uptake capacity of the wheat/buckwheat crops. Consequently, nitrate levels in leachate remained unacceptably high throughout the monitoring period 1974 to 1976 even though average annual NO_3-N levels declined from year to year. Concentrations of NO_3-N were also unacceptably high from the NPK control treatment on both soils. Even leachate from non-fertilized control lysimeters had NO_3-N values which exceeded 10.0 mg/l. These data illustrate the difficulty in controlling nitrogen leakage when low nitrogen uptake crops such as wheat are grown. If nitrate pollution of groundwater as a result of sludge application to soil becomes a major factor limiting sludge application rates, similar restrictions in application of commercial NPK

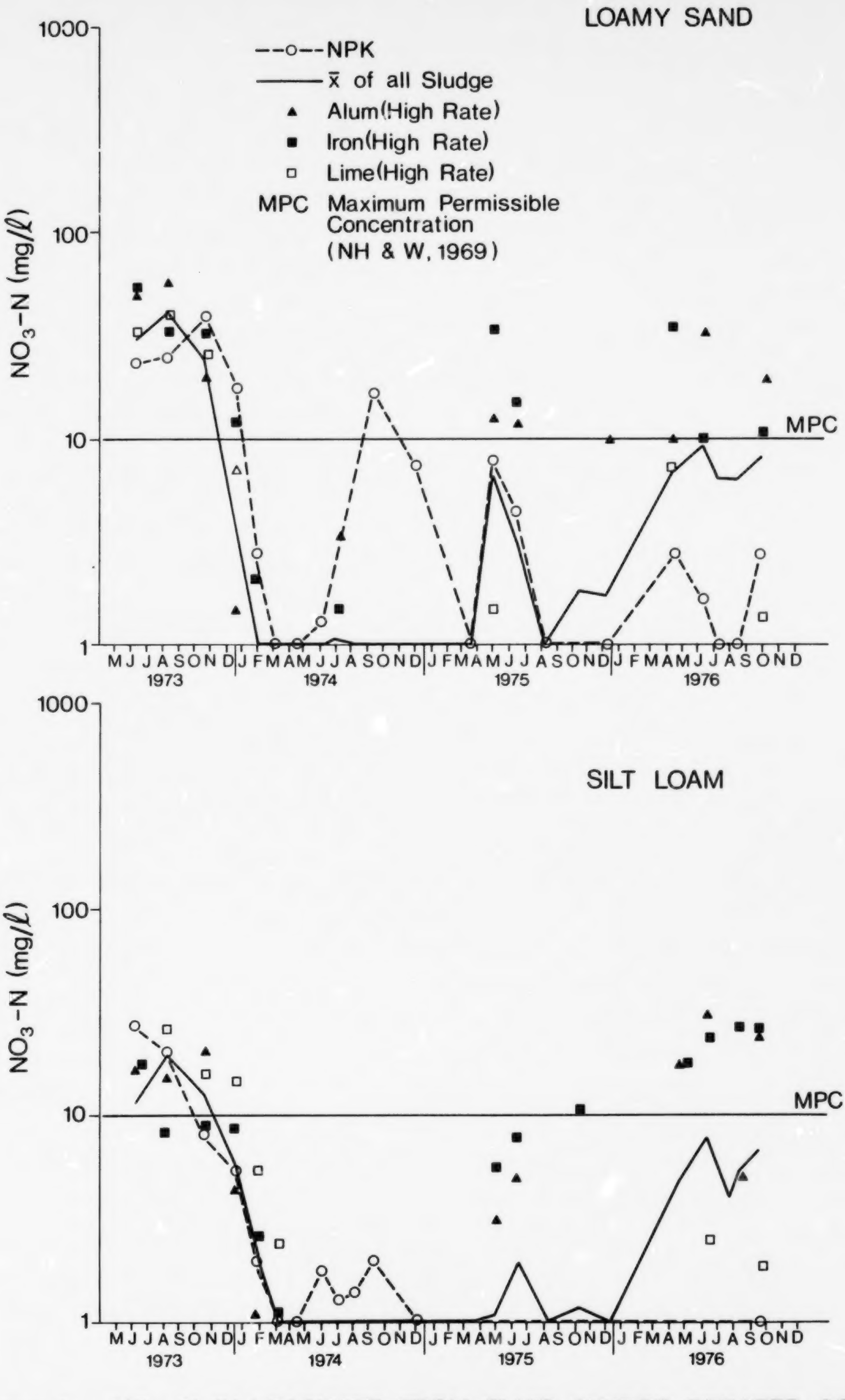


Figure 21 NO$_3$-N IN LEACHATE FROM FLUID SLUDGE TREATED SOILS

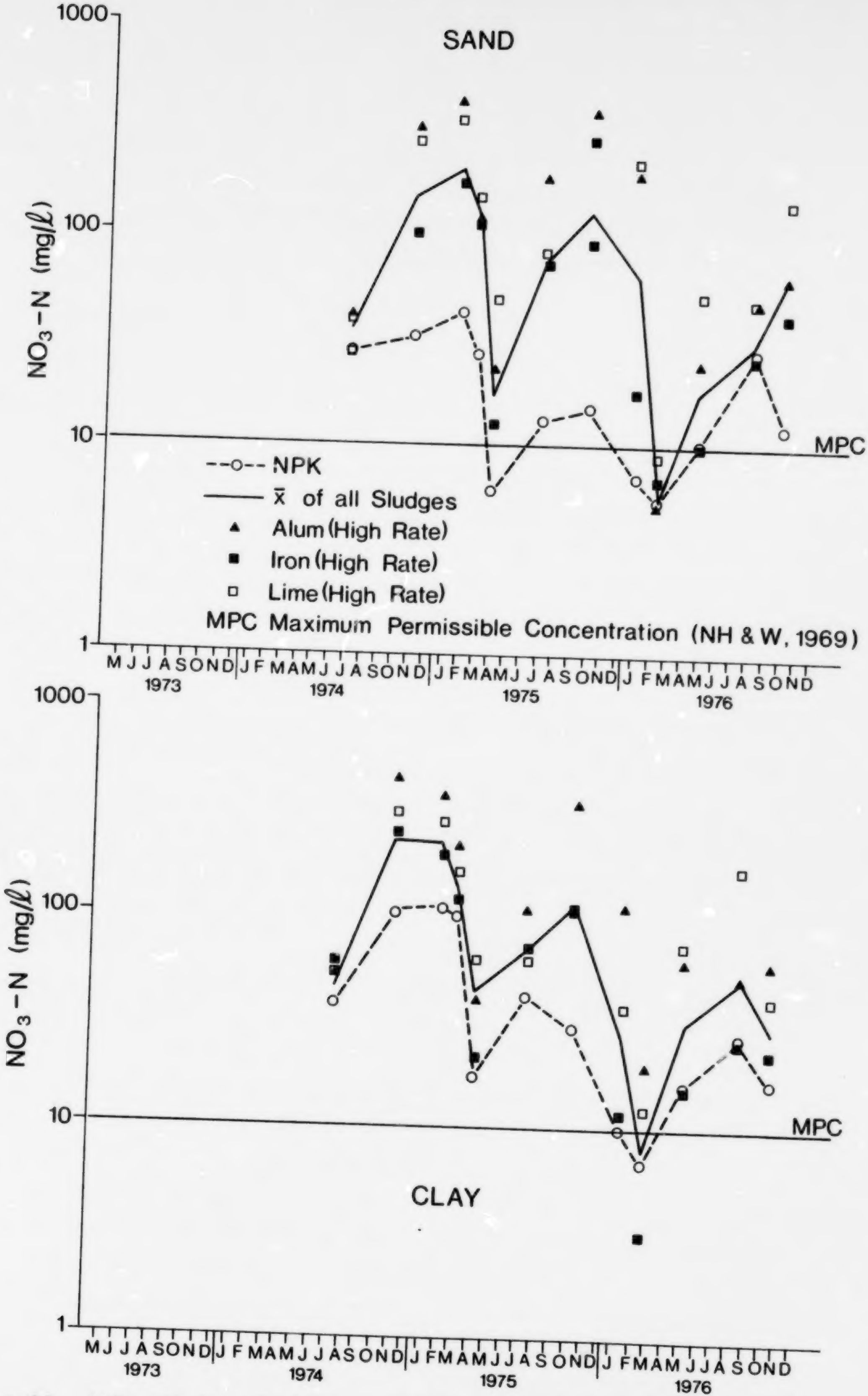
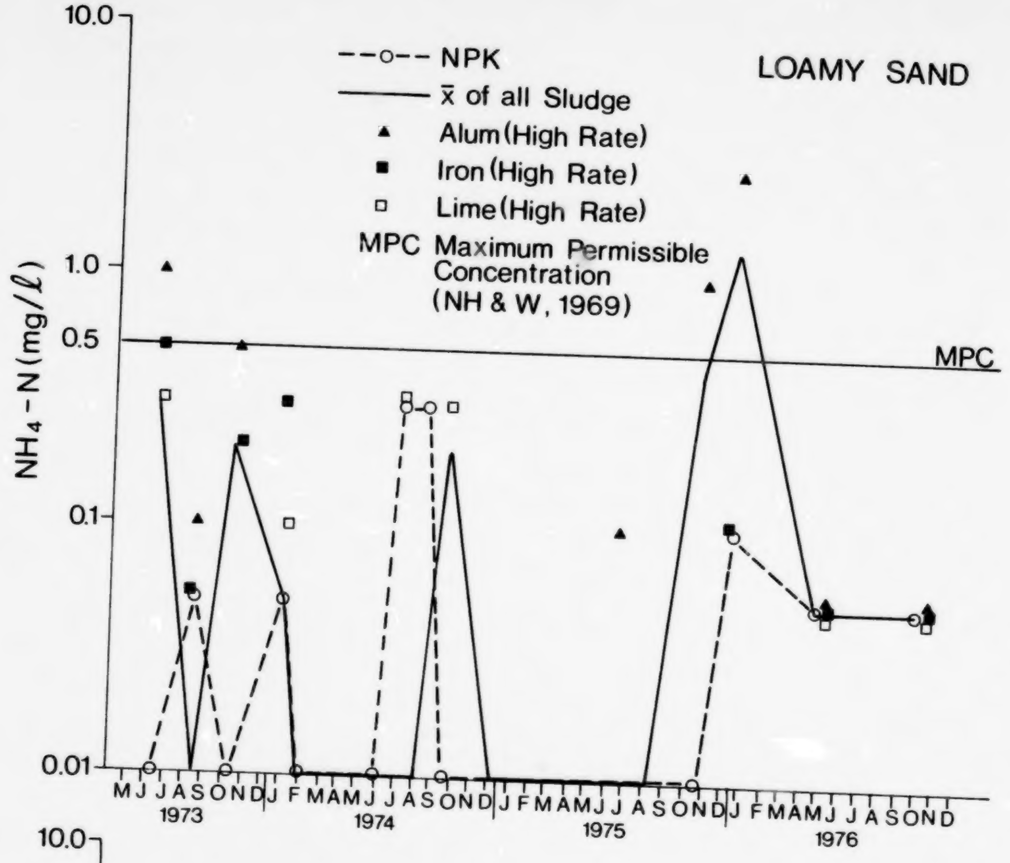


Figure 22 NO_3-N IN LEACHATE FROM AIR-DRIED SLUDGE TREATED SOILS fertilizers may be expected. Background information concerning the seasonal variability of NO_3-N concentrations in groundwater where no fertilizer is applied would be of benefit in evaluating the impact of sludge loadings on groundwater quality.

Figures 23 and 24 show changes in NH_4-N concentrations with time in leachate from fluid and air-dried sludge amended soils, respectively. The maximum permissible NH_4-N concentration (0.50 mg/l) was exceeded on loamy sand in 1975 (alum sludge high rate) but declined in 1976 below the MPC. Ammonium ion concentrations in leachate from silt loam were below the MPC (mean of all sludge treatments) throughout the monitoring period (1973 to 1976) and closely approximated NH_4-N concentrations from NPK control treatment.

When air-dried sludges were applied (Figure 24), NH_4-N values above the MPC were observed for sandy soil during the first year only. Clay soil NH_4-N leachate concentration never exceeded the MPC indicating rapid conversion of organic nitrogen to NO_3-N.

Figures 25 and 26 show changes in soluble P concentrations with time in leachate from fluid and air-dried sludge amended soils, respectively. Phosphorus in leachate is of particular concern in view of the ongoing effort, at great expense, to remove it from sewage effluent in the Great Lakes basin area. The existing effluent objective is 1.0 mg P/l, while Canadian drinking water standards (NH&W, 1969) require a concentration of less than 0.10 mg P/l to avoid interference with treatment processes or algal growth in surface waters. After initial stabilization of the fluid sludge orchard grass lysimeters in 1973, soluble P concentrations in leachate from both loamy sand and silt loam generally remained below the MPC until the winter of 1975/76 when concentrations increased above 1.0 mg/l, particularly from alum sludge at the highest loading rate. Concentration of P declined during the 1976 growing season but, after the final grass harvest, P in leachate from both soils increased again. A similar trend towards soluble P concentration in excess of 1.0 mg/l from both air-dried sludge and NPK treatments on sandy soil (Figure 26) was observed after the wheat harvest. Soluble P in leachate from clay soils never exceeded 0.7 mg/l for any treatment.



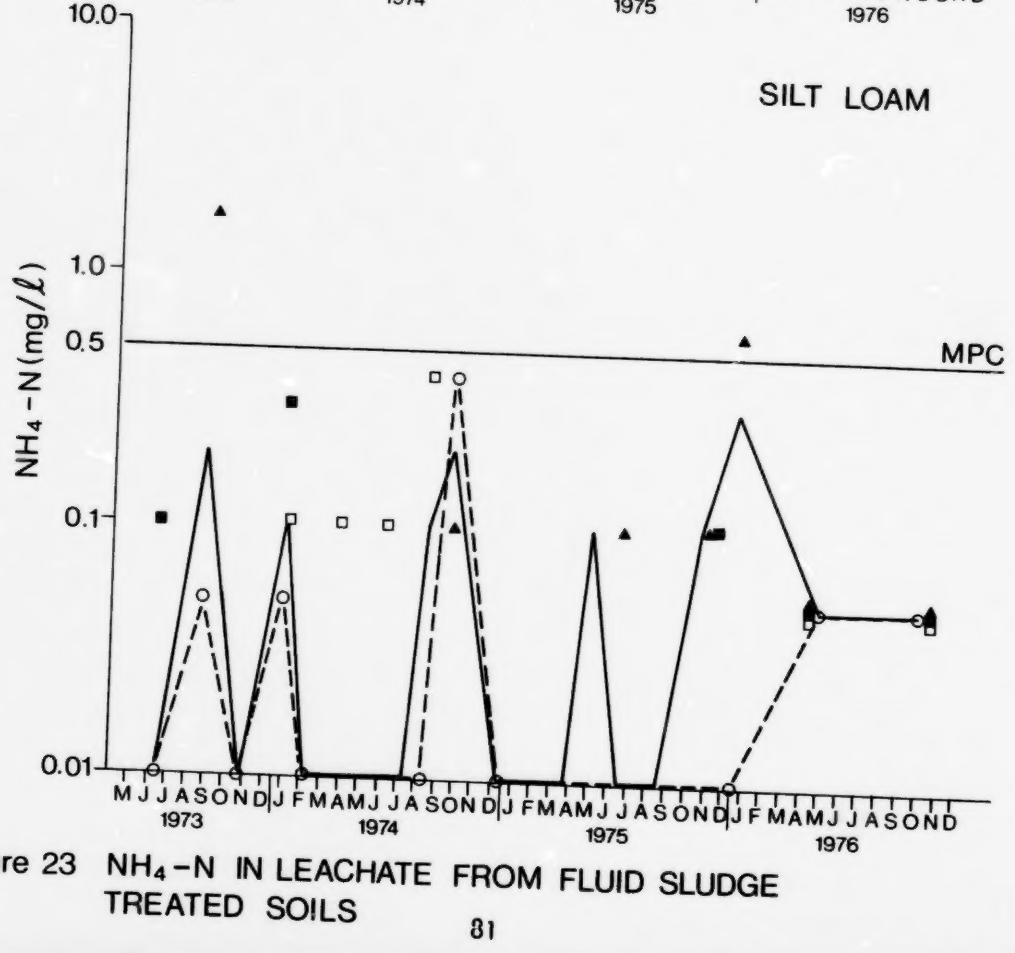


Figure 23 NH₄-N IN LEACHATE FROM FLUID SLUDGE TREATED SOILS

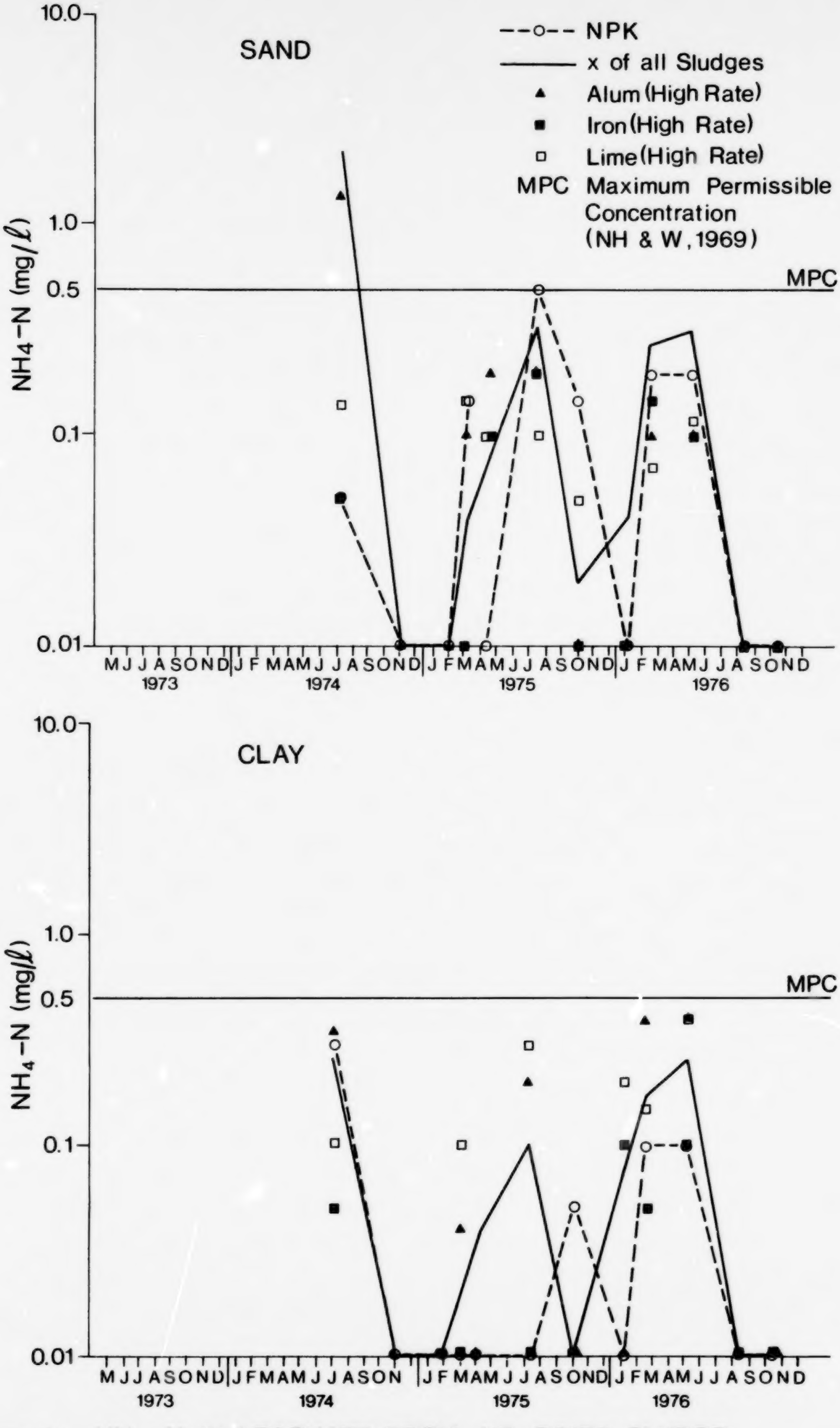


Figure 24 NH$_4$-N IN LEACHATE FROM AIR-DRIED SLUDGE TREATED SOILS

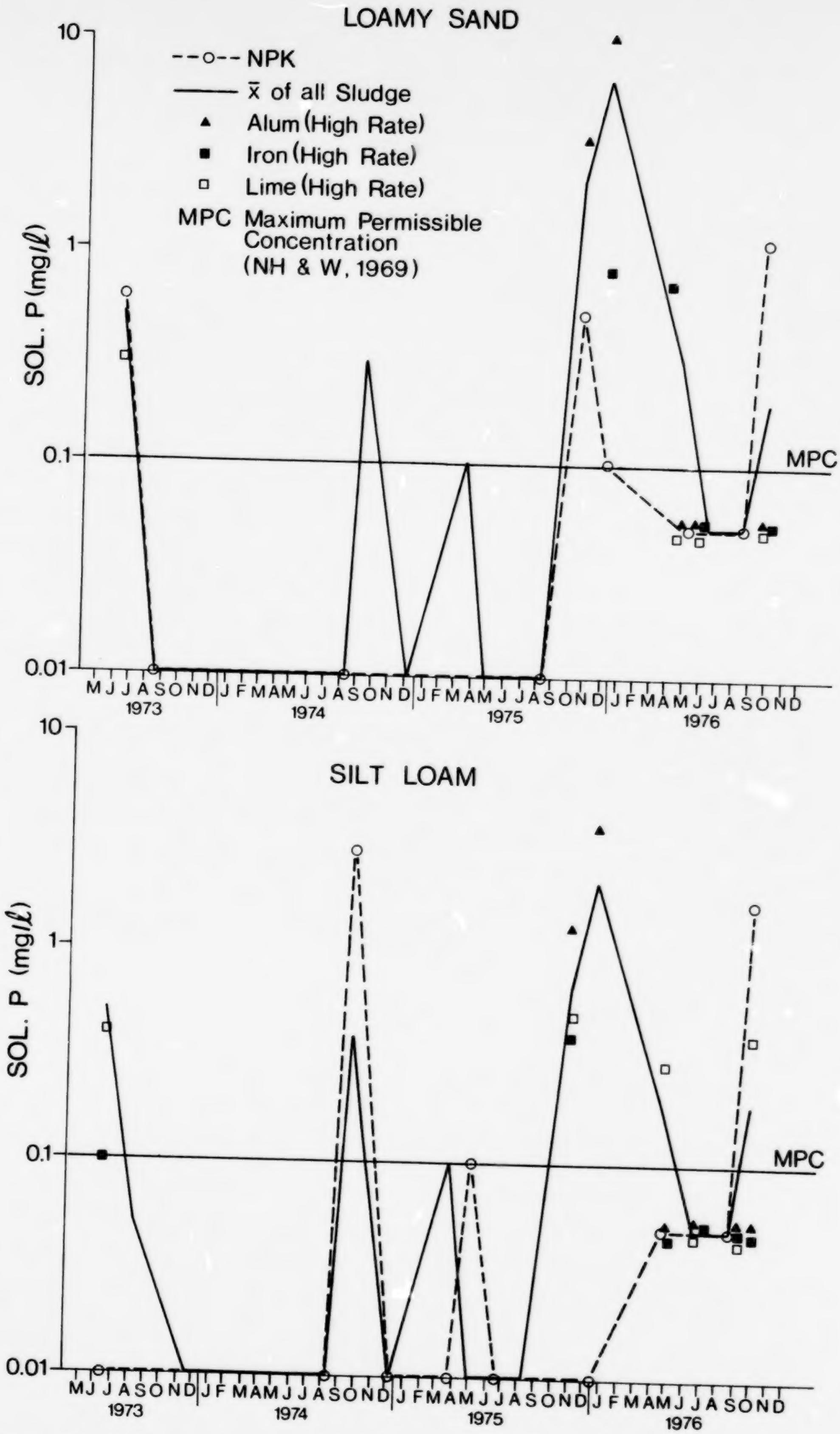


Figure 25 SOLUBLE P IN LEACHATE FROM FLUID SLUDGE TREATED SOILS

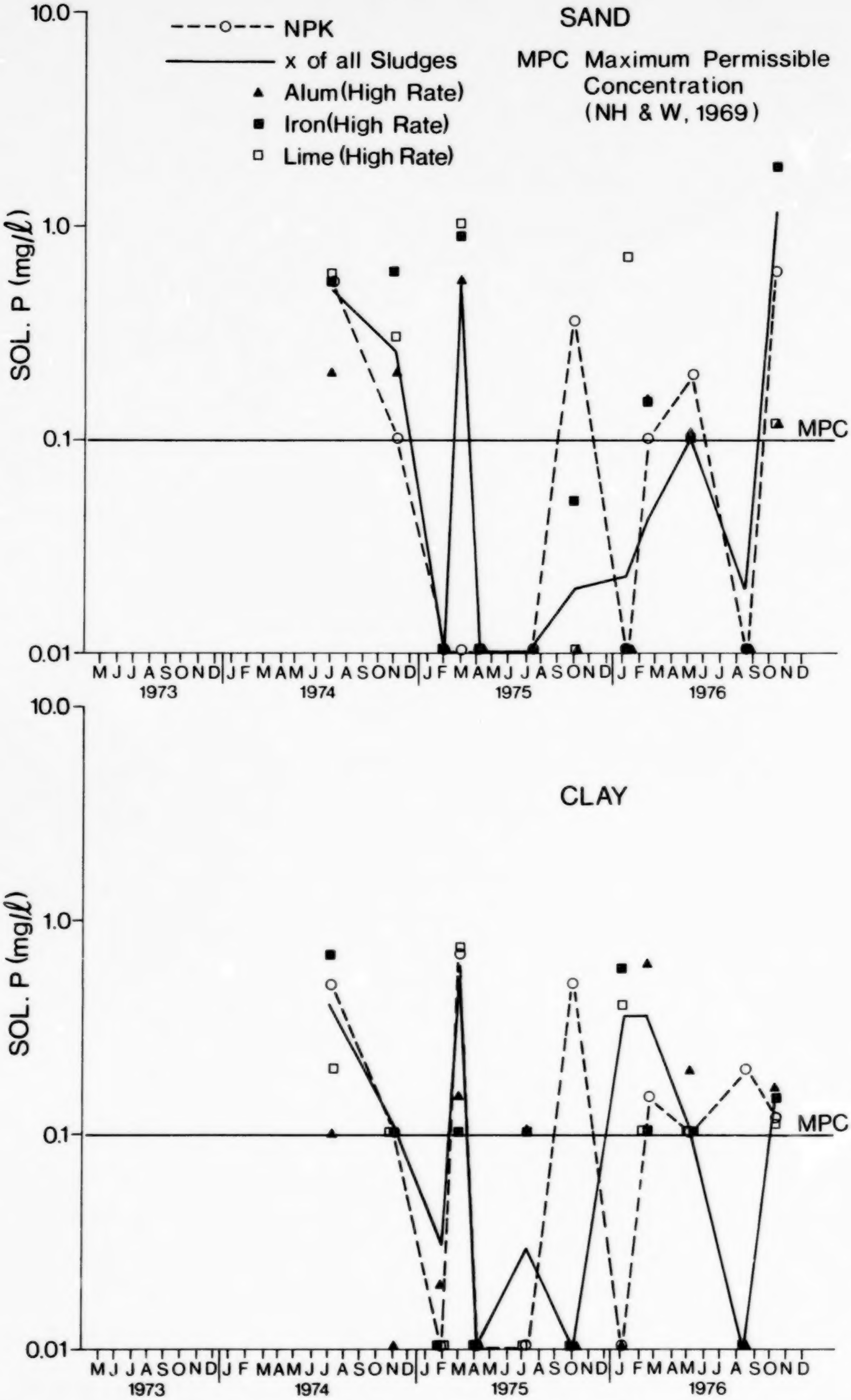


Figure 26 SOLUBLE P IN LEACHATE FROM AIR-DRIED SLUDGE TREATED SOILS

Figures 27 and 28 show changes in TOC concentration with time in leachate from fluid and air-dried sludges, respectively. Although the actual TOC/BOD ratio must be determined experimentally, 15.0 mg/l was used to define the MPC for this parameter, as a safety factor until the toxicity of the organic components contributing to the TOC value have been identified. TOC in leachate from the orchard grass soil systems was low in 1973, but exceeded the MPC in the fall of 1974. TOC values declined below the MPC in 1975, but increased again in the summer of 1976, with values in excess of 50 mg/l recorded at the highest loading rates.

Static bioassay toxicity tests were run on leachate collected in September, 1976 using Daphnia pulex. The results were negative when undiluted samples were tested.

Bioassay toxicity tests were also run on leachate from the air-dried sludge experiments with similar results. In view of these findings, the carbon chloroform extract (CCE) test will be substituted in future for TOC to provide a more sensitive indicator of potentially deleterious organic substances in leachate.

Other Leachate Constituents. No changes with time for soil type, sludge type, or rate, were observed for values of pH, Fe, Mg and K in leachate from the fluid sludge-orchard grass system.

Concentrations of Na, Ni and Pb decreased with time with no significant differences due to soil, sludge type, or rate.

The following parameters all showed slight increases with time, although maximum values were within the relevant water quality acceptable limits (maximum values in brackets).

Conductivity (990 micromho/cm^2); Cl (61 mg/l); SO_4 (240 mg/l); Ca (165 mg/l); Mn (0.54 mg/l); and Al (0.12 mg/l).

Somewhat higher maximum leachate concentrations were observed for the various constituents from the air-dried sludge-wheat system. Maximum values were within acceptable drinking or irrigation water limits as follows: Cl (122 mg/l); SO_4 (300 mg/l); Ca (470 mg/l); Mg (122 mg/l); Na (52 mg/l); K (5.3 mg/l); Fe (0.16 mg/l); Mn (0.54 mg/l); and Al (0.48 mg/l).

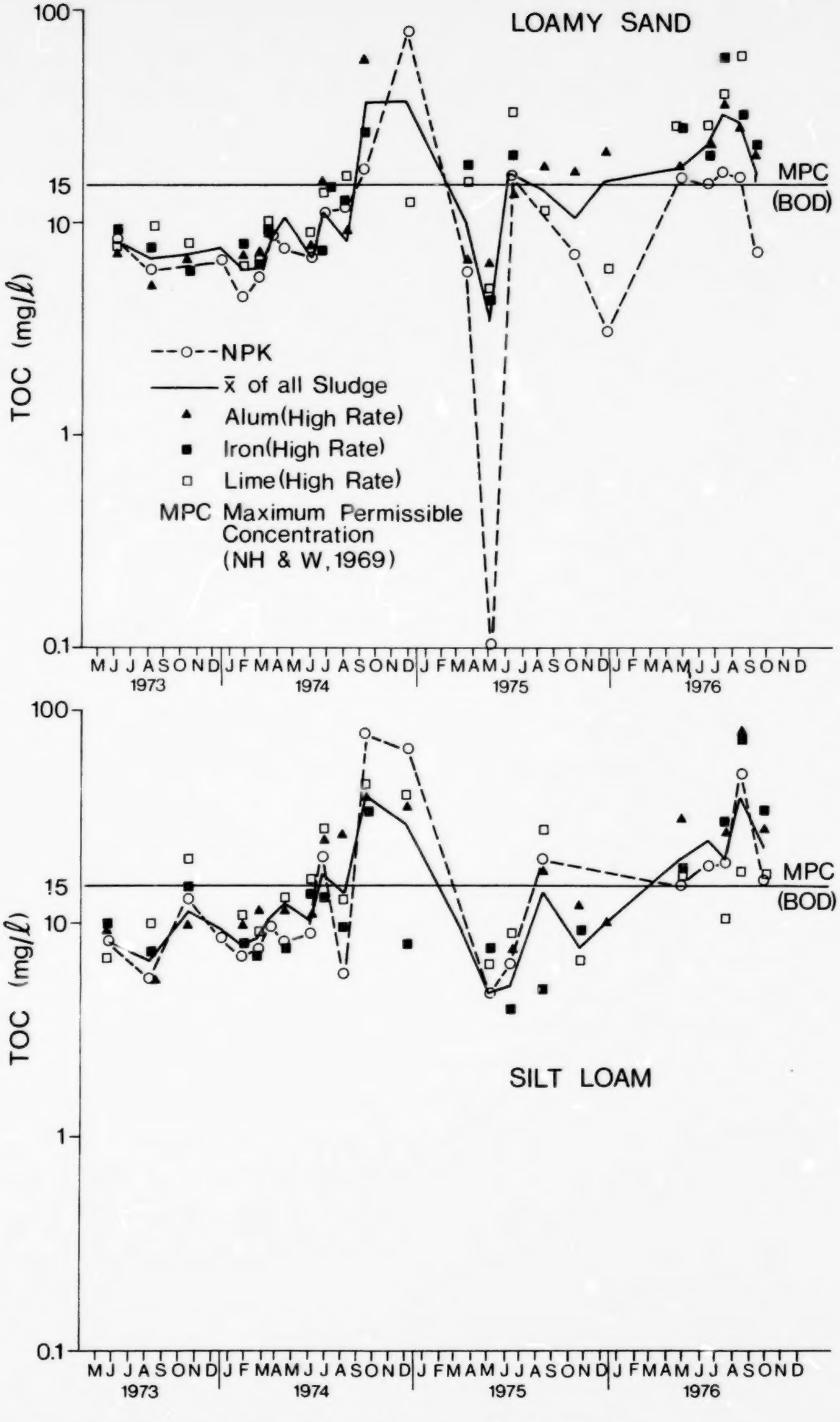


Figure 27 TOC IN LEACHATE FROM FLUID SLUDGE TREATED SOILS

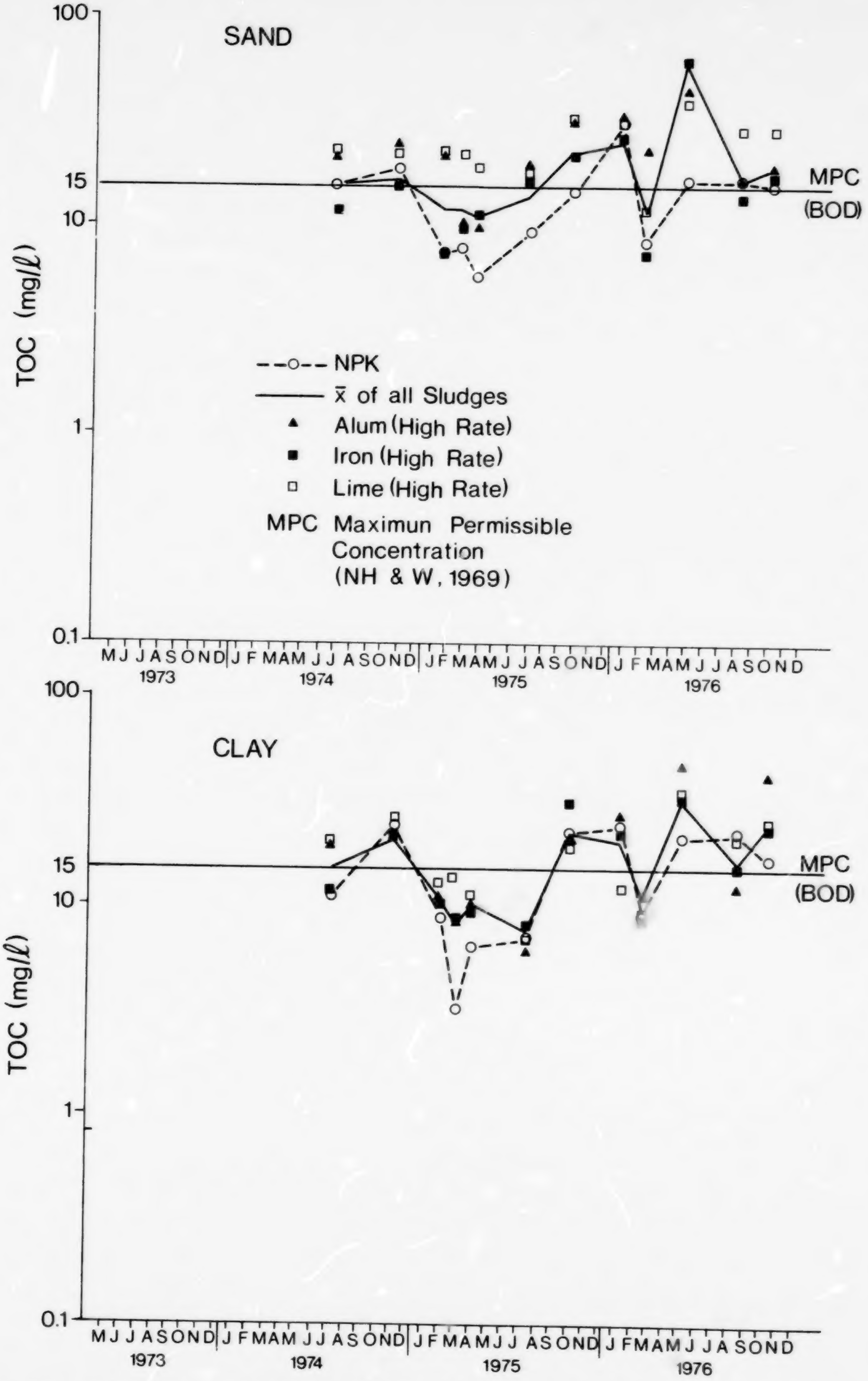


Figure 28 TOC IN LEACHATE FROM AIR-DRIED SLUDGE TREATED SOILS

5.4.2.1 Leachate concentration deviation from controls. Concentrations of the various constituents of concern in leachate may be contributed in part by natural soil processes independent of fertilizer application. Similarly, the contribution of N, P and other nutrients from commercial fertilizers applied at the recommended rates must be known to properly assess the potential long term hazards of sludge application to farmland in relation to groundwater quality.

When groundwater background levels of a particular chemical constituent exceeded the water quality standard MPC, application of any fertilizer, commercial or sludge, would have to be carefully controlled to avoid exacerbating an already "bad" situation. Concentration of a particular nutrient in leachate would have to be reduced by limiting the application rate if either the crop uptake capacity or soil "storage" capacity were exceeded.

Even if the groundwater background level of a particular nutrient was acceptable, application of NPK fertilizer at the recommended rate might increase the concentration in leachate up to or beyond the acceptable water quality MPC. The effect on leachate quality of applying sludges at varying loading rates should, therefore, be compared to the NPK effect.

There are several possible ways to relate maximum allowable sludge application rates to acceptable leachate quality.

1. Accept drinking or irrigation water quality criteria as the MPC standard for leachate.
2. Accept non-fertilized control leachate values as "standard".
3. Accept leachate quality from NPK fertilizer applied at the agronomically recommended rate as "standard".

If the first criterion was adopted, neither NPK nor sludge treatments would be allowed where groundwater background concentrations exceeded the relevant water quality MPC.

If the second criterion was adopted, leachate values above the control level would require reduction or cessation of any fertility treatment or change in agronomic practice (crop selection) to reduce leakage to acceptable levels.

If the third criterion was adopted, leachate values from most sludge treatments would be acceptable.

Guidelines for sludge application to farmland do not address this problem directly. The Ontario provisional guidelines (1976) assume implicitly that, if recommended rates of fertilizer are applied, groundwater quality will be protected but no provision or standards are quoted to enable quantification of this assumption. The difficulty and expense of adequately monitoring either leachate or groundwater from sludged farms is, undoubtedly, the major reason this question has not been adequately addressed to date.

In subsequent paragraphs, leachate nutrient concentrations from sludge application at the highest rate are compared with both NPK and non-fertilized controls. These comparisons illustrate some of the problems one faces in deciding which of the three above mentioned groundwater protection strategies should be pursued.

Nitrate (NO_3-N). When fluid sludge was applied cumulatively to orchard grass at nitrogen loading rates three times higher than NPK, no adverse effects were noted during the first two years (Figure 29). During the third and fourth years, however, the residual organic nitrogen mineralization effect increased in both soils. Lower plant uptake of N from loamy sand resulted in leachate from alum and iron sludges exceeding NPK average annual concentrations by more than 10.0 mg/l.

This increasing nitrate concentration trend is similar whether NPK or non-fertilized controls are used as the basis of comparison. NO_3-N average annual values after the first year never exceeded 4.7 mg/l for either NPK or non-fertilizer controls. It is noteworthy that lime sludge application even at the highest rate (unlike alum and iron sludges) did not result in unacceptable NO_3-N leachate levels in either soil during four years of monitoring. Further research is required to determine whether this differential nitrate formation is due to inhibition of soil nitrifying bacteria or to accelerated gaseous nitrogen losses as NH_3 or N_2.

A very different situation occurs when air-dried sludge is applied in a single annual heavy application (1974) to sand and clay

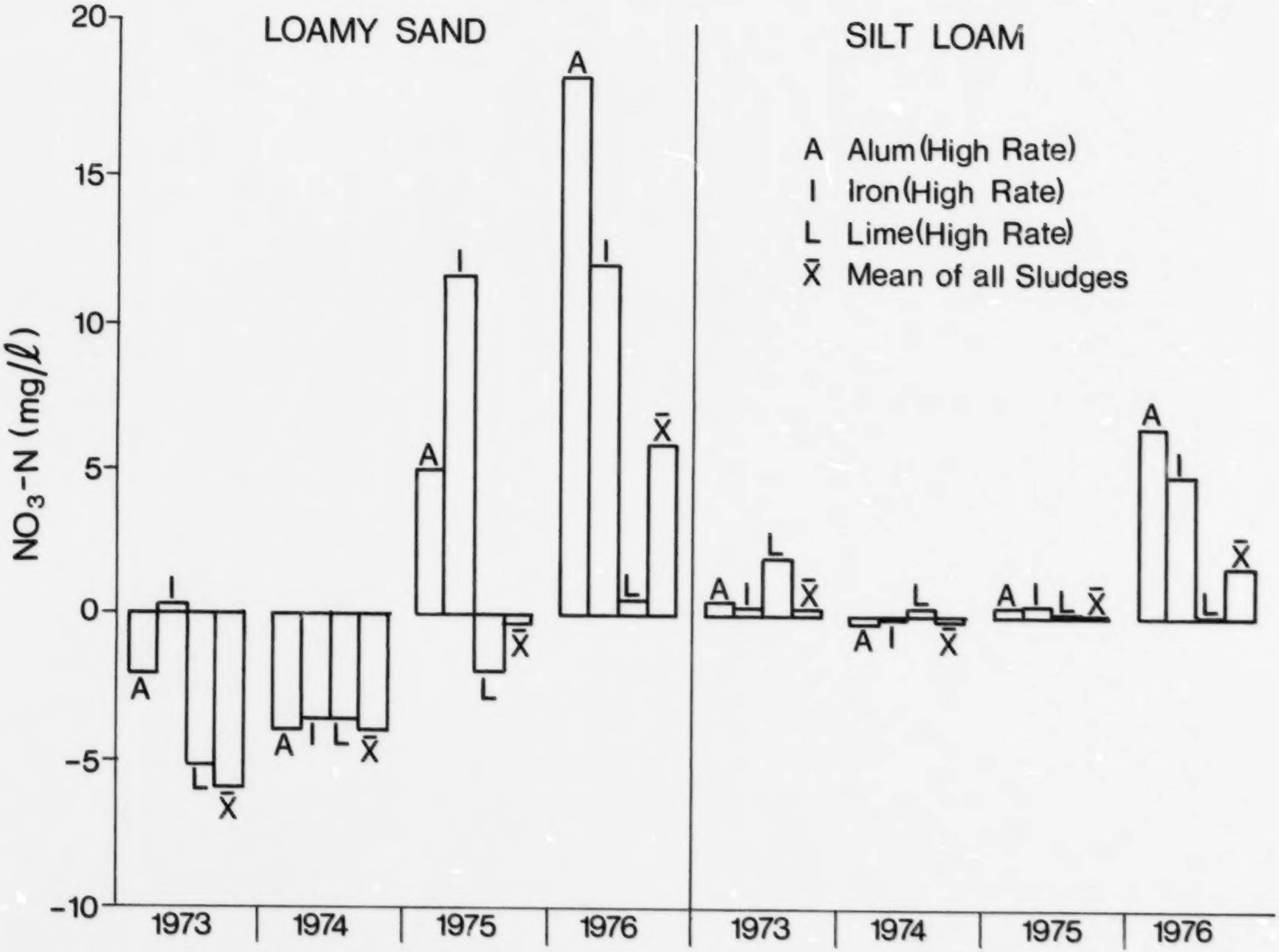


Figure 29 NO_3-N DEVIATION FROM NPK CONTROL (LEACHATE)

(Fluid Sludge - Orchard Grass)

soils (Figure 30). Unacceptably high NO_3-N levels in leachate, exceeding both NPK and non-NPK control by >200 mg/l, appear during the first year. In the second year these deviations from control decline, but remain unacceptably high (>10 mg/l) two years after sludge has ceased to be applied. Undoubtedly, heavy loadings of air-dried sludge to either light or heavy textured soils, growing a relatively low nitrogen uptake crop (wheat), are a serious danger to groundwater quality several years after the initial loading event. If prevention of nitrate pollution of groundwater is a major objective of a well managed sludge to soils system, great care must be exercised in selection of sludge loading rate and crops.

Ammonium (NH_4-N). A comparison of NH_4-N levels in leachate from fluid sludges (high rate) and NPK treatments to orchard grass (Figure 31) reveals that with the exception of alum sludge, deviations are minimal on both soils (<0.1 mg/l). NH_4-N is almost invariably converted to NO_3-N in groundwater. This potential increment to the groundwater background NO_3-N concentration is not considered at this time to be a cause for concern even from the "worst case" alum sludge. Concentrations of NH_4-N from both NPK and non-fertilized controls (both soils) were virtually identical (<0.1 mg/l) and unchanging during the monitoring period 1973 to 1976.

Ammonium ion concentration in leachate from the air-dried sludge treatments (Figure 32) was frequently less than from either NPK or non-fertilized control. Sludge treatments at all loading rates showed mean deviations from control of less than 0.1 mg/l. Consequently NH_4-N pollution of groundwater from this sludge source (unlike NO_3-N pollution) is not considered to be a major cause for concern at this time.

Soluble P. During four years of monitoring leachate from the fluid sludge-orchard grass experiment for soluble P (Figure 33) only one treatment (alum sludge at high rate on loamy sand) indicated P transport through the soil profile. The leachate phosphorus concentration exceeded 1.0 mg P/l. Soluble P from the non-fertilized control increased from 0.1 mg/l (both soils) in 1973 and 1974 to 0.70 mg/l (loamy sand) in 1976.

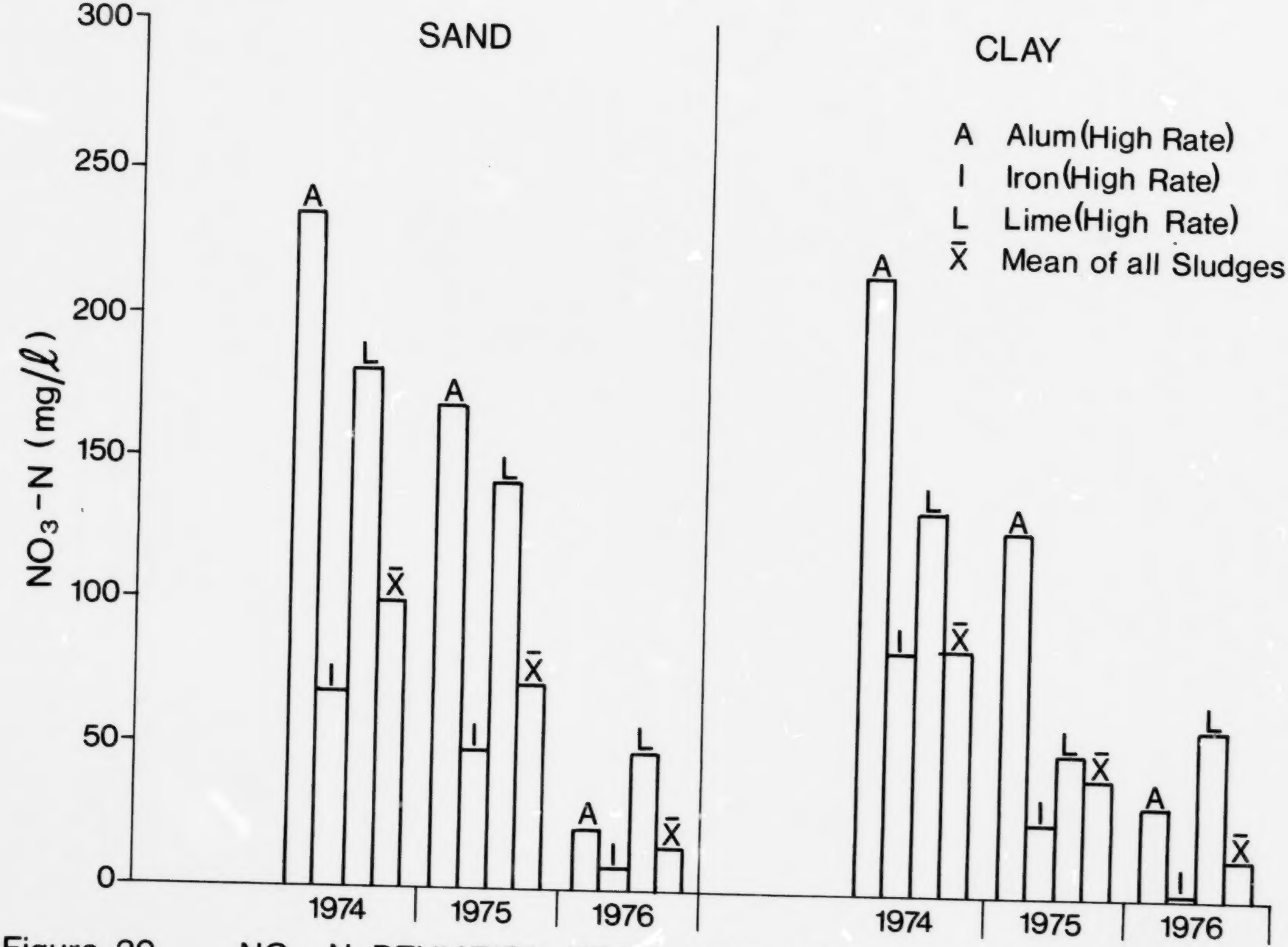


Figure 30 NO_3-N DEVIATION FROM NPK CONTROL (LEACHATE)

(Air-Dried Sludge-Wheat)

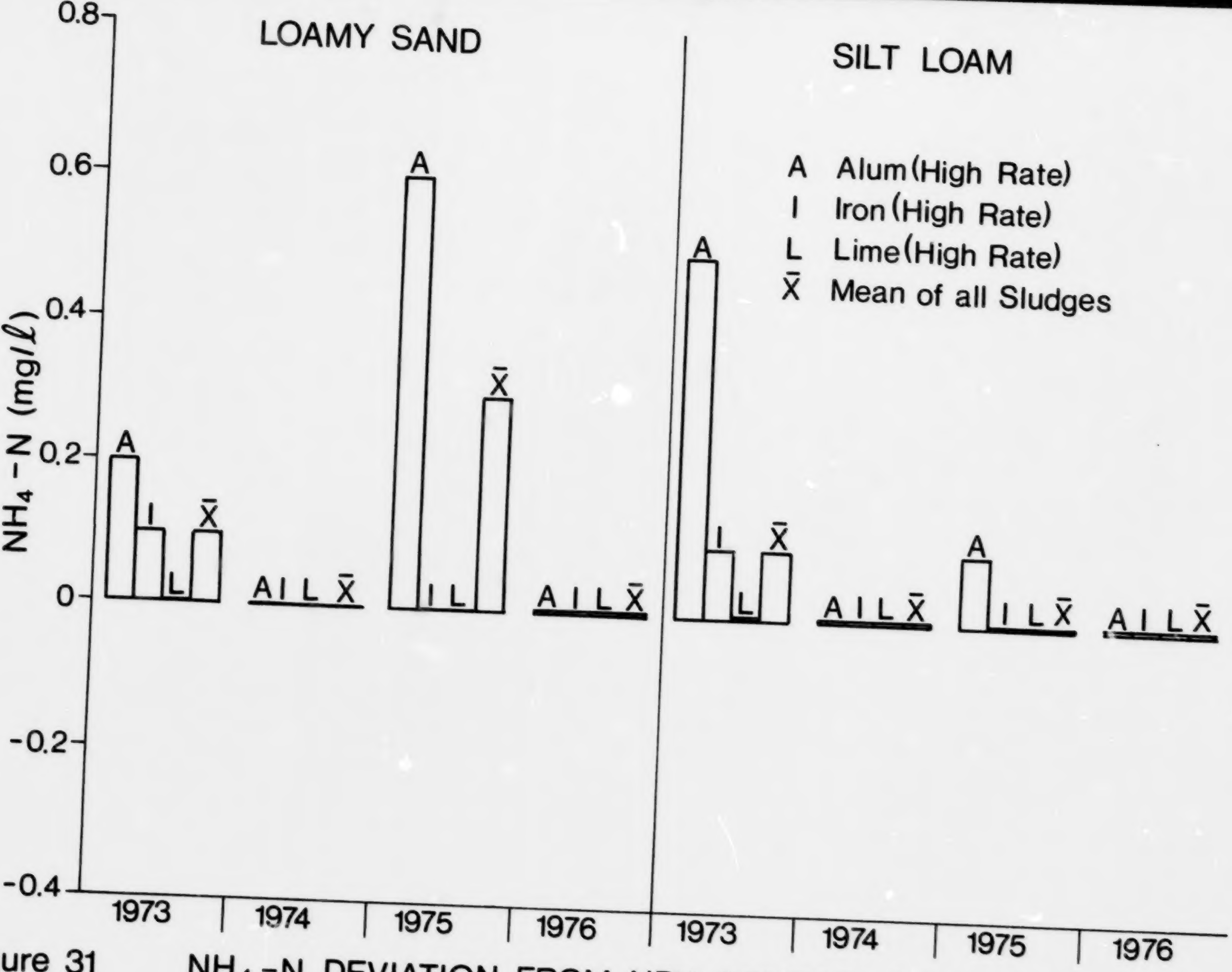


Figure 31 NH_4-N DEVIATION FROM NPK CONTROL (LEACHATE)

(Fluid Sludge-Orchard Grass)

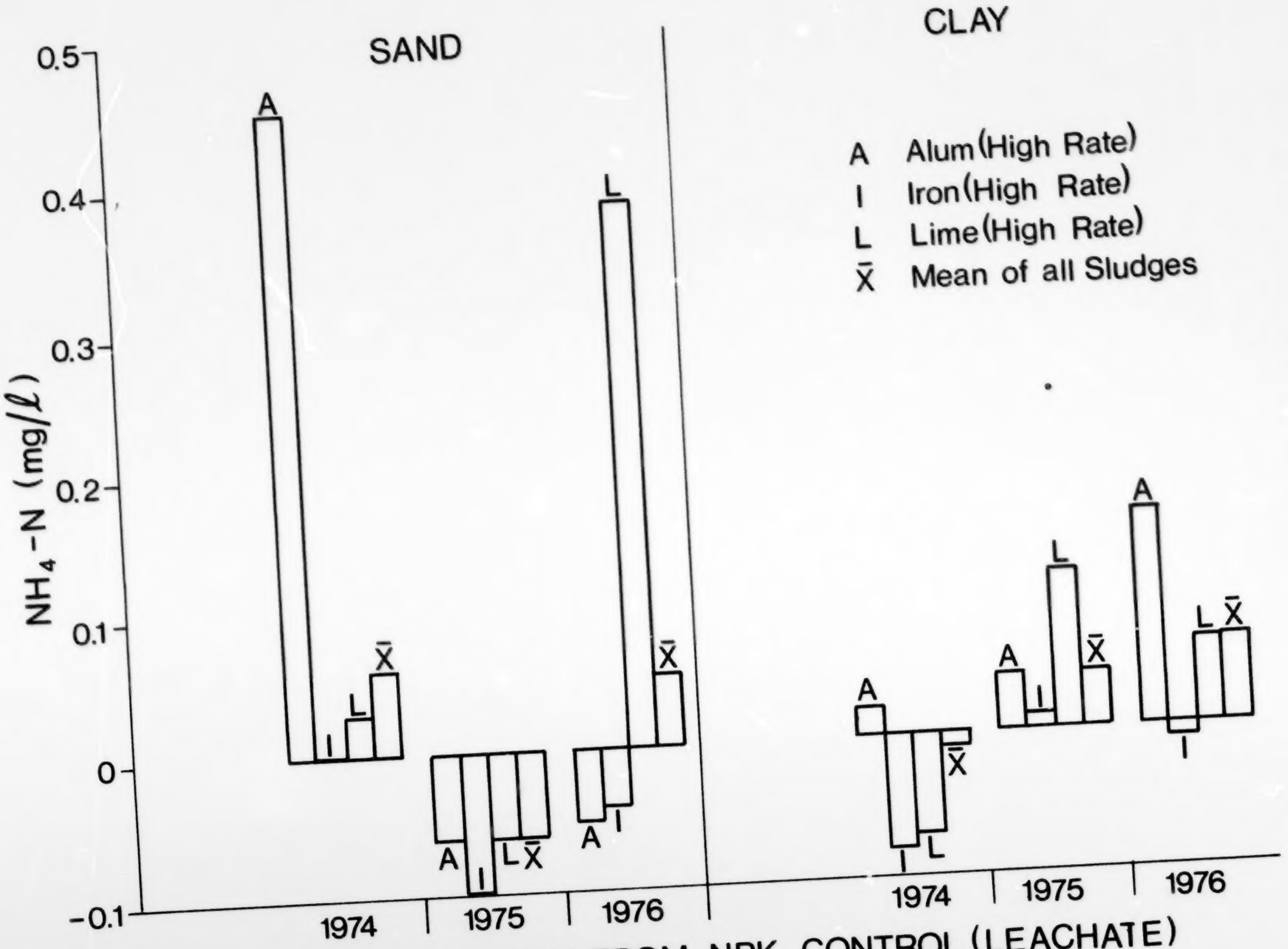


Figure 32 NH$_4$-N DEVIATION FROM NPK CONTROL (LEACHATE)
(Air-Dried Sludge-Wheat)

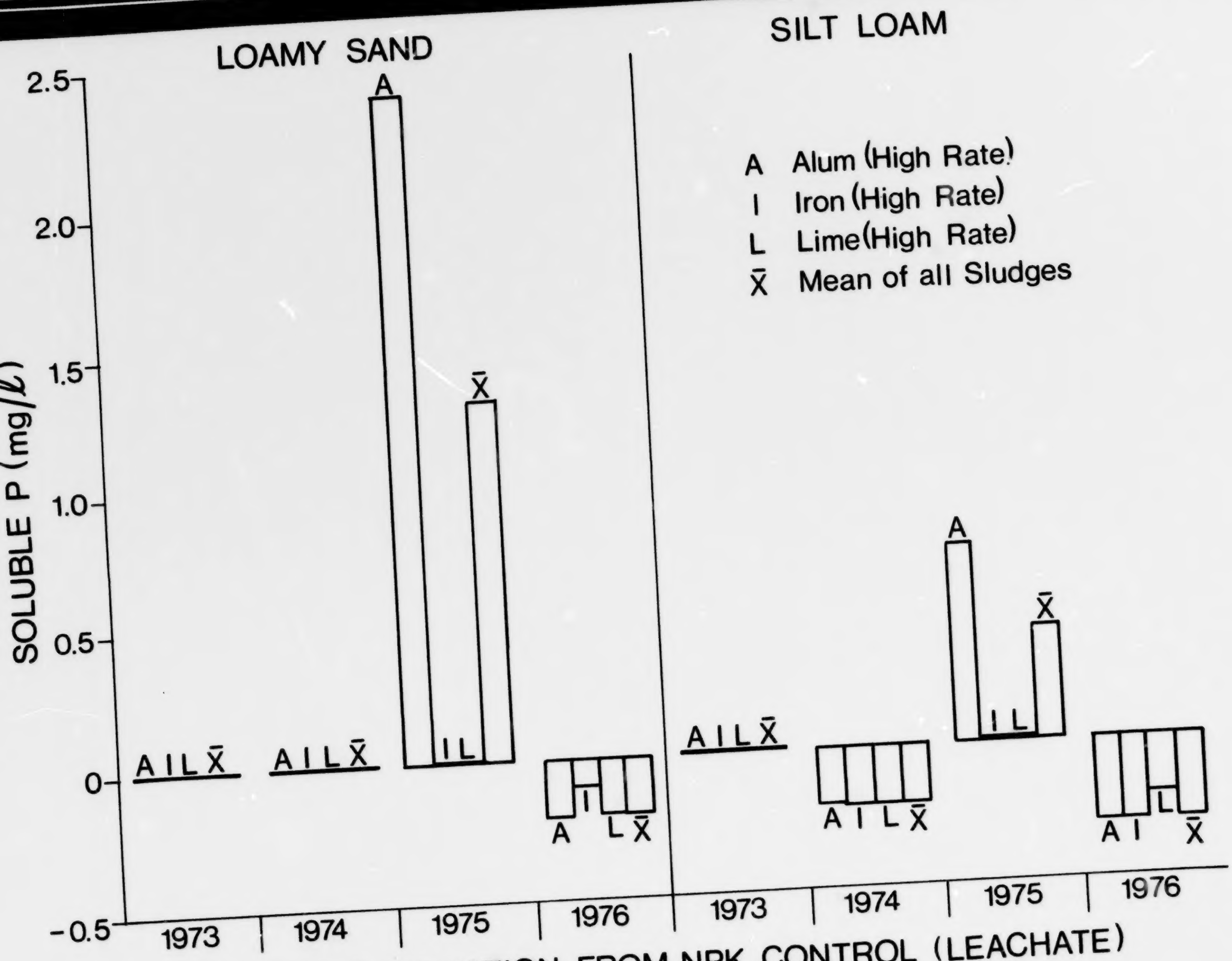


Figure 33 SOLUBLE P DEVIATION FROM NPK CONTROL (LEACHATE)

(Fluid Sludge-Orchard Grass)

Soluble P from NPK increased from 0.10 mg/l during 1973, 1974 and 1975 to 0.40 mg/l from silt loam in 1976. These concentrations approach or exceed the recommended drinking water MPC of 0.5 mg/l. The 1976 high rate sludge treatment leachate results from both soils are less than either NPK or non-fertilized control values. Under the conditions of this study, the leaching of phosphorus into the groundwater table does not appear to be a problem. A possible exception could be alum sludge at the high application rate.

Figure 34 illustrates the deviation with time of soluble P from air-dried sludge treatments compared to non-fertilized control results. On sandy soil the variability of P in leachate is very high, the sludge treatment values being 0.7 mg/l lower than those of the non-fertilized control. On clay soil, the variability of soluble P in leachate is less than on sandy soil, but no trend towards buildup with time can be discerned. Pollution of groundwater with soluble P above the natural background levels cannot, therefore, be considered a serious potential threat at this time.

Total Organic Carbon. Buildup of organic carbon compounds above natural background levels in groundwater is usually a consequence of man's activities. Total organic carbon was chosen as a measure of organic carbon in leachate because of concern for the potential toxicity or carcinogenicity of unknown organic carbon compounds in drinking water and the need for a rapid and simple analytical method.

Figure 35 illustrates the deviation of TOC in leachate from fluid sludge (high rate) treatments compared to the NPK control values (6 to 24 mg/l). These deviations are even greater if non-fertilized control values are used. Many of these "humic" organic compounds are not readily biodegradable. Equivalent BOD values, as determined by parallel laboratory analysis of selected leachate samples, were approximately half the reported TOC values. A significant increase in sludge treatment TOC concentrations compared to NPK control was observed in 1975 (loamy sand) and in 1976 (both soils). The need for more specific information concerning the chemical composition of the organic carbon compounds contributing to this increase has led to the recommendation to

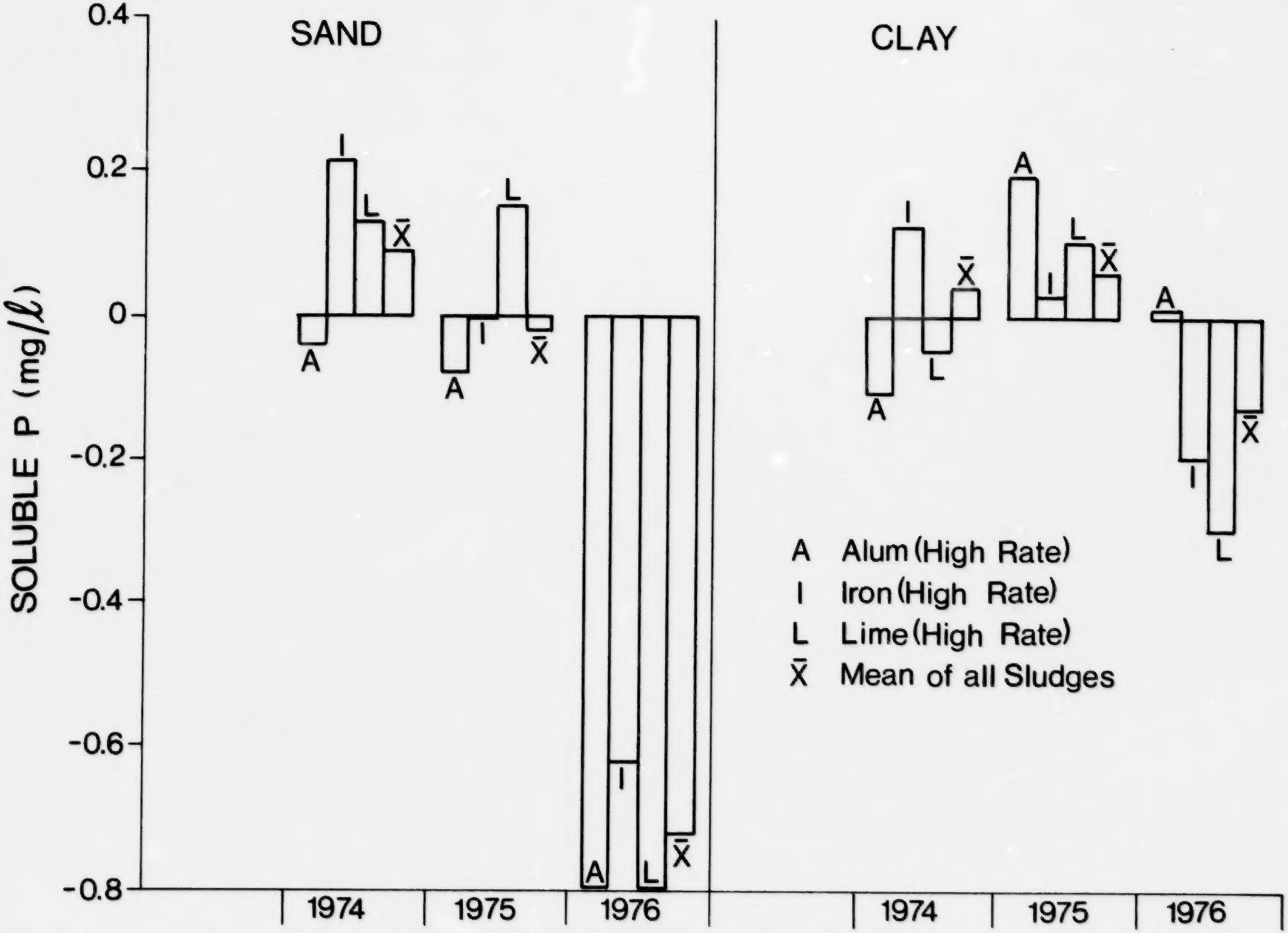


Figure 34 SOLUBLE P DEVIATION FROM NON-NPK CONTROL (LEACHATE)

(Air-Dried Sludge-Wheat)

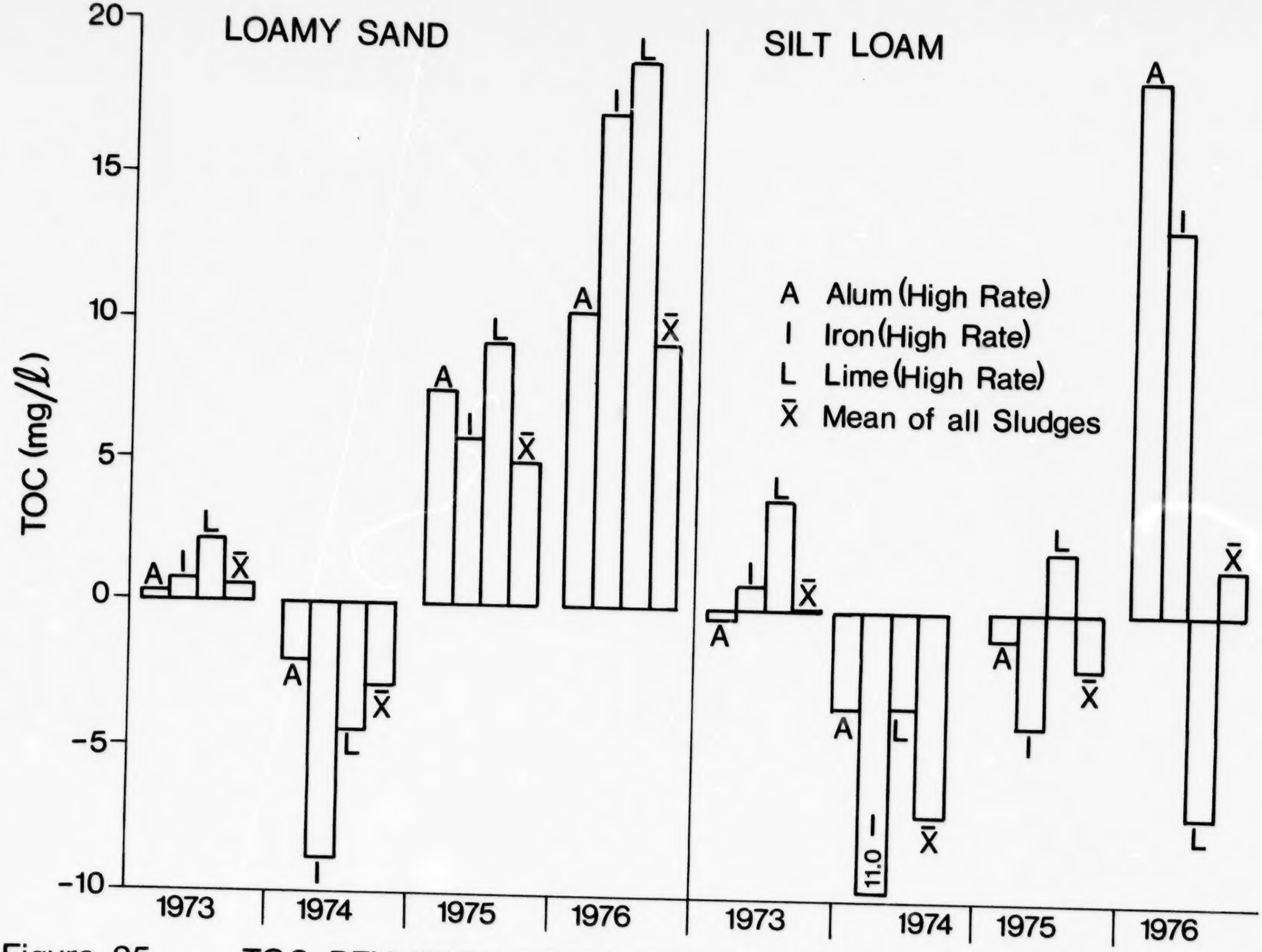


Figure 35 TOC DEVIATION FROM NPK CONTROL (LEACHATE)
(Fluid Sludge-Orchard Grass)

analyze carbon chloroform extract (CCE) in 1977. The present drinking water standard for CCE is 0.15 mg/l (OMOE, 1974). If this concentration is exceeded, identification of the specific chemical compounds (e.g., pesticides) contributing to the CCE will be undertaken, by gas chromatography.

Some interesting differences are noted between the TOC in leachate from the air-dried sludge experiments and the fluid sludge experiment results. TOC values for NPK controls from both sand and clay soils were relatively constant with time, ranging between 12 to 17 mg/l. The TOC values in leachate from the non-NPK controls, however, were higher than the NPK values, ranging between 16 to 21 mg/l on sandy soil and between 42 to 55 mg/l from clay soils. The application of air-dried sludge to clay soil reduced the TOC leakage to groundwater by approximately 40 mg/l, compared to non-NPK control values. The deviation from non-NPK control is becoming more positive annually, from -6 mg/l in 1974 to +6 mg/l in 1976 on sandy soil, and from -39 mg/l in 1974 to -21 mg/l in 1976 on clay soil (Figure 36). These results illustrate the complex nature of the various organic-inorganic interactions that occur in soils both naturally and as a result of sludge addition.

5.4.3 Leachate cumulative losses and recovery efficiency

When all the data concerning leachate quality trends, including deviation from control values have been reviewed, NO_3-N remains the major constituent of concern, under the conditions of these experiments. Heavy metals and other nutrient losses, in terms of quantity leached per hectare of soil farmed per year, are several orders of magnitude less than nitrate losses (<1 kg/ha/yr).

Figure 37 shows trends, with time, in total quantities of NO_3-N leached from loamy sand and silt loam soils for the various fluid sludge and control treatments. After the first (non-typical) year, when soils in the lysimeters were undergoing a period of stabilization, NO_3-N leakage from the NPK treatment averaged 4 kg/ha/yr from loamy sand and 2 kg/ha/yr from silt loam. As 300 to 500 kg N/ha/yr were applied to the NPK control lysimeters annually, these very low losses represent less

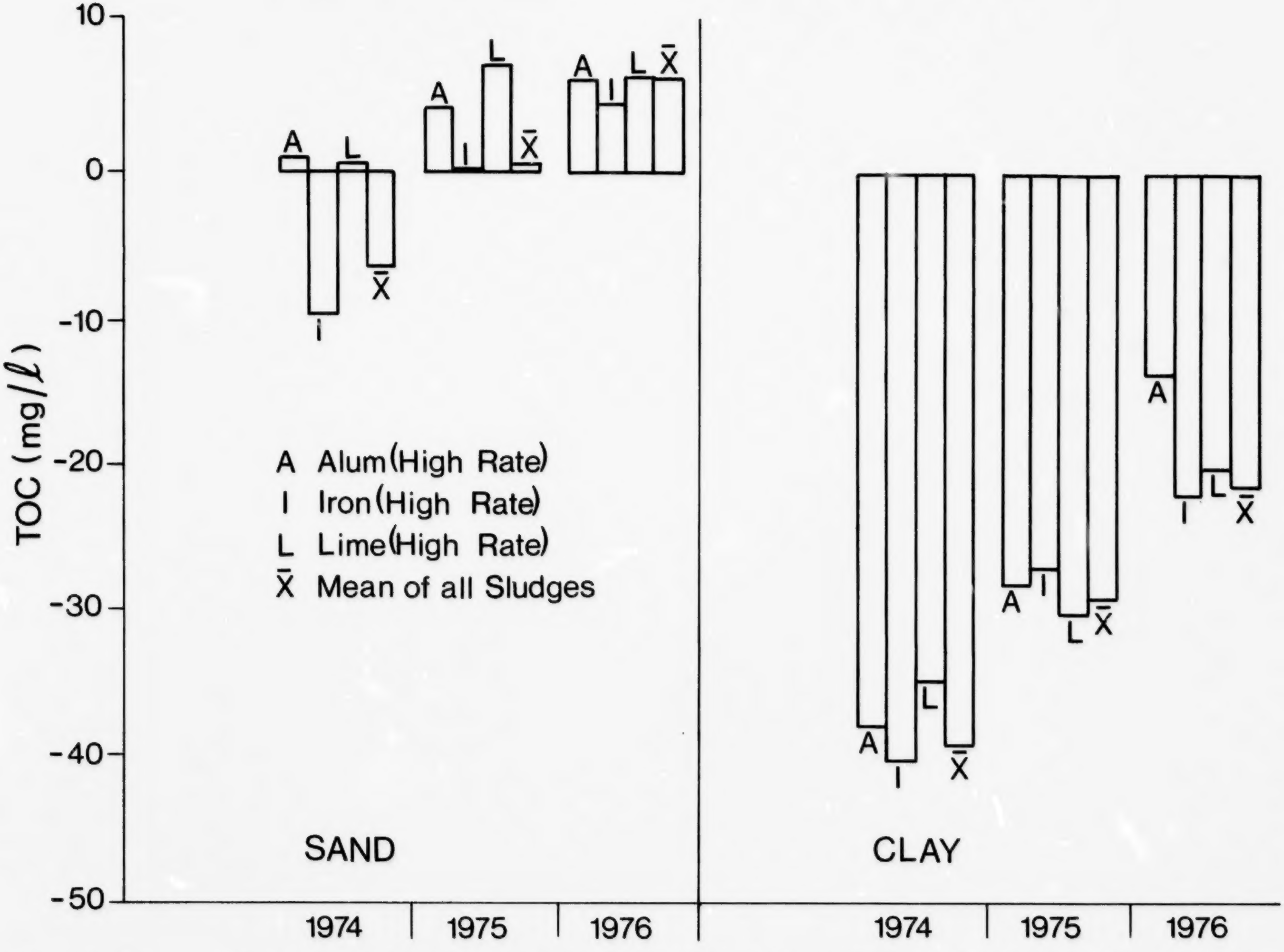


Figure 36 TOC DEVIATION FROM NON-NPK CONTROL (LEACHATE)
(Air-Dried Sludge-Wheat)

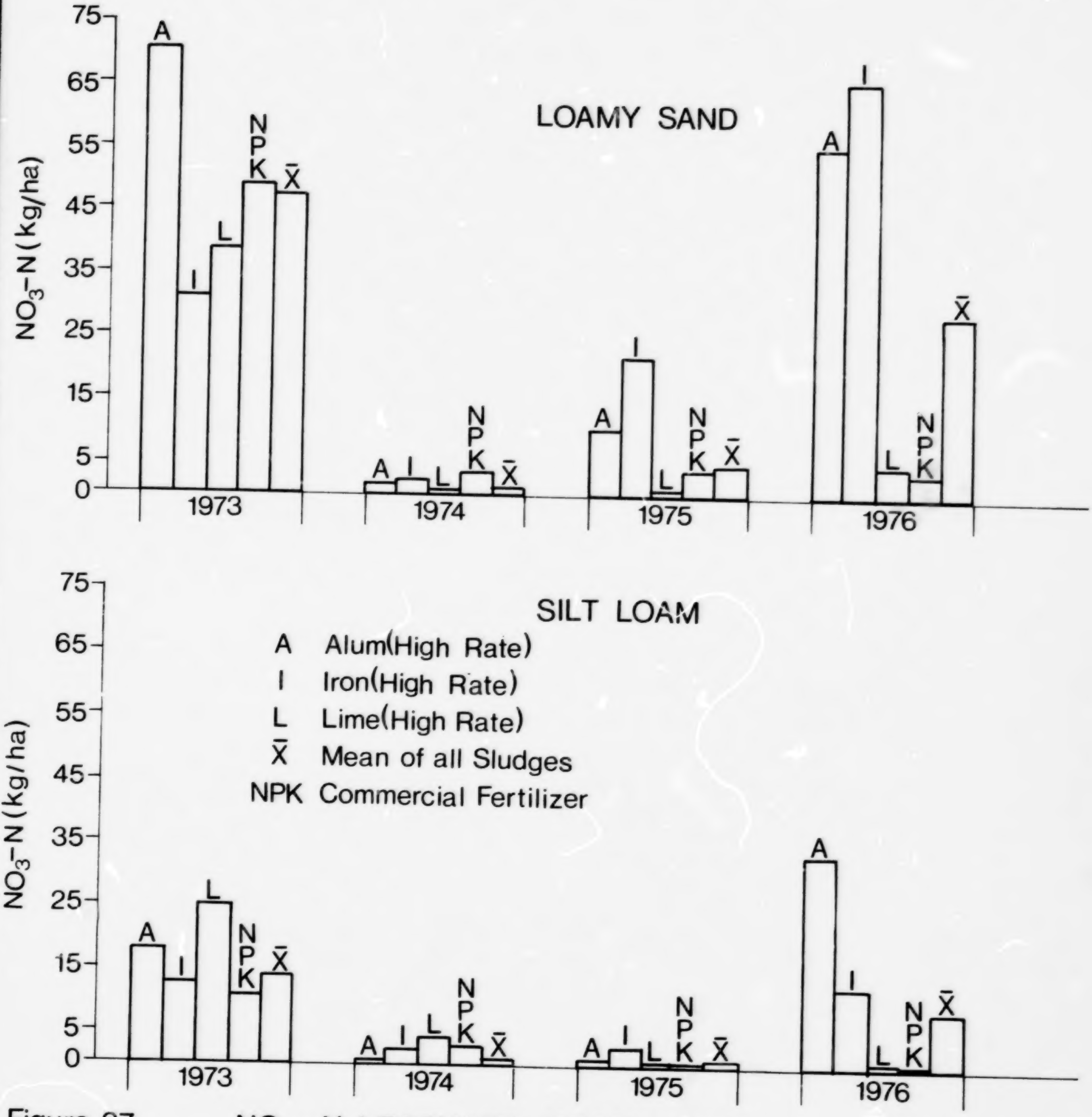


Figure 37 NO_3-N LEACHATE ANNUAL LOSS
(Fluid Sludge-Orchard Grass)

than 1% of the nitrogen applied. In view of the large quantities of organic nitrogen contained in sewage sludges, which undergo nitrification after completion of the growing season, any treatment causing less than 10 kg/ha/yr NO_3-N leakage has been defined for the purposes of this study as "acceptable".

In 1974, all nine sludge treatments on both soils, including the heaviest application rates, produced an "acceptable" amount of NO_3-N leakage. During the third year (1975), NO_3-N leakage remained acceptable on silt loam, but the coarser textured loamy sand leaked unacceptable amounts of NO_3-N from the highest rates of alum and iron sludge application. By the end of the fourth year (1976) alum and iron sludges on both soils were producing unacceptable NO_3-N losses (up to 65 kg/ha/yr at the high rate). For reasons which are not yet clear, the lime sludge application continued to produce an "acceptable" rate of NO_3-N leakage even after four consecutive years of sludge application. One possible explanation of this difference in leachate nitrate formation may be that lime sludges undergo increased NH_3 volatilization at the time of application. Soil profile analyses in 1977 should indicate whether soils receiving lime sludges contain significantly less soil N than do soils receiving either alum or iron sludges at equivalent TKN loading rates.

TKN application rates for iron, alum and lime air-dried sludges applied to sand and clay soils were not constant, unlike the fluid sludge experiments. To compare the relative acceptability of the three sludge types with regard to NO_3-N leakage, the absolute amounts of nitrate leached were corrected by calculating the relative amounts leached per 10^3 kg TKN/ha applied. Figure 38 shows that lime sludges produce far less NO_3-N in leachate than do alum or iron sludges at equivalent loading rates (as was the case with fluid sludges). Alum (high rate) sludge produces the highest amount of NO_3-N leached from both soils. By the end of the third year (1976) NO_3-N lost per 10^3 kg/TKN applied was approaching acceptable levels (10 kg NO_3-N/ha/yr) for all three sludge types on sandy soils and for iron and lime sludges on clay soil. Re-application of air-dried sludge at the end of the 1976 growing season may reverse the trend towards acceptable NO_3-N leachate losses

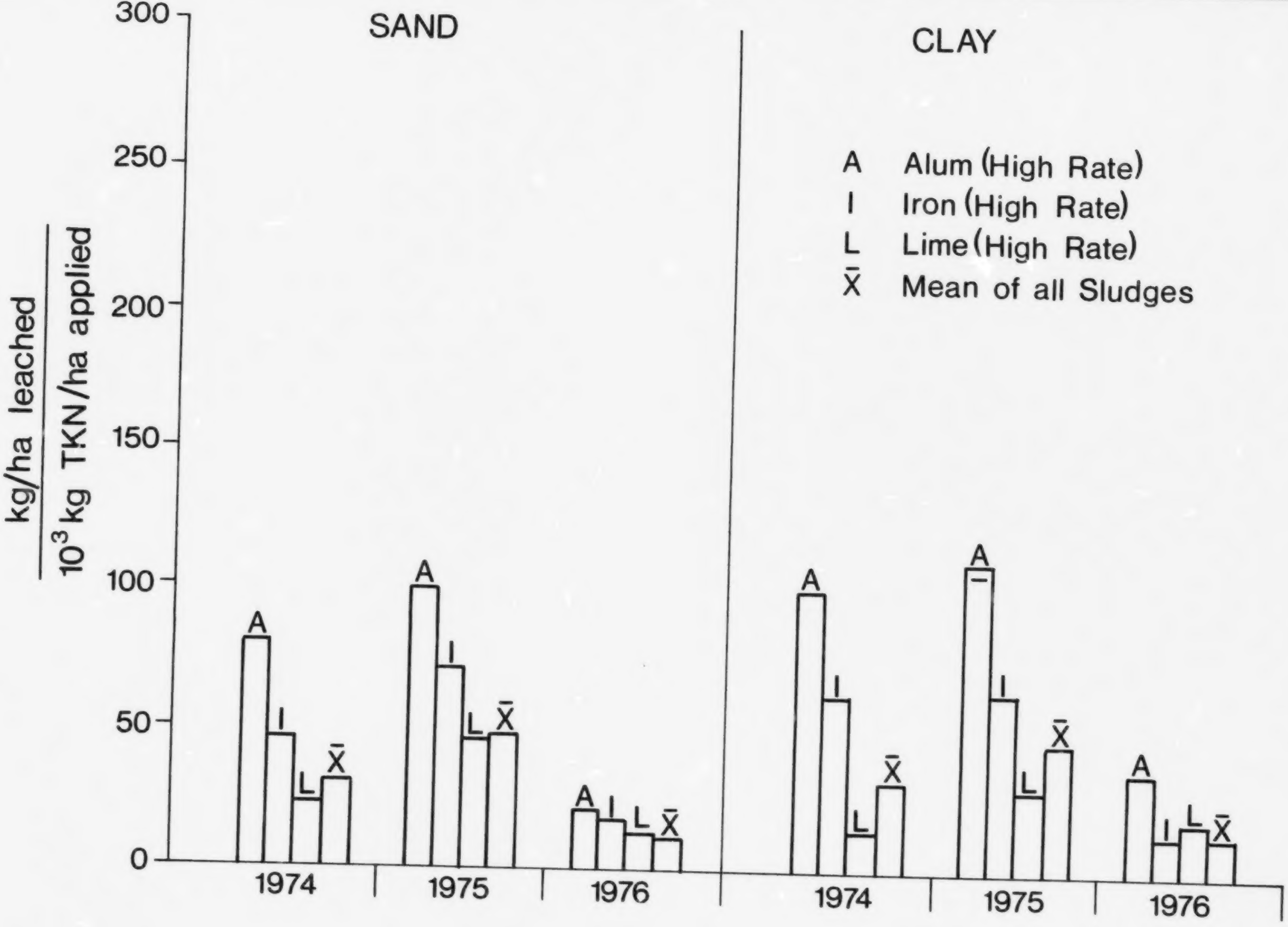


Figure 38 NO_3-N LEACHATE ANNUAL LOSS
(Air-Dried Sludge-Wheat)

for several more years, until a new equilibrium is reached in the soil between nitrogen applied and nitrogen lost by leaching, crop uptake, volatilization or denitrification.

Table 31 summarizes the maximum cumulative leachate losses (worst case) of nutrients and heavy metals from both fluid and air-dried sludge amended soils during the period 1973 to 1976.

TABLE 31. MAXIMUM CUMULATIVE LOSSES OF NUTRIENTS AND HEAVY METALS (kg/ha) IN LEACHATE FROM SLUDGE AMENDED SOILS AT THE HIGHEST LOADING RATE

Constituent	Fluid Sludge (Orchard Grass) (1973-76)		Air-Dried Sludge (Wheat) (1974-76)	
	Loamy Sand	Silt Loam	Sand	Clay
N	210	103	1153	1162
P	23	4	2	2
Zn	0.28	0.25	0.31	0.20
Cu	0.31	0.38	0.31	0.20
Ni	0.19	0.18	0.08	0.04
Cr	0.06	0.04	0.20	0.07
Pb	0.21	0.22	0.21	0.10
Cd	0.03	0.02	0.03	0.02

Nitrogen losses from loamy sand were double those from silt loam, while losses from sand and clay soils receiving air-dried sludge were more than ten times the silt loam losses on an average annual basis. Metal losses never exceeded 0.10 kg/ha/yr for any particular constituent. All six metals combined leached less than 0.4 kg/ha/yr. Combined metal losses from loamy sand and silt loam were virtually identical. Cumulative losses from sandy soil of all six metals monitored (1.14 kg/ha) were nearly double the losses of these metals from clay soil (0.63 kg/ha) during the period 1974 to 1976. The unusually high P loss from loamy sand was due to a single sludge source - alum, at the highest loading rate.

The ratios of the maximum sludge treatment losses versus "normal" losses of nutrients and metals from the NPK control treatment during the same monitoring period are summarized in Table 32.

TABLE 32. RATIO OF MAXIMUM CUMULATIVE LEACHATE LOSSES (kg/ha) FROM SLUDGE AMENDED SOILS AT THE HIGHEST LOADING RATE[1] vs NPK CONTROL LOSSES[2]

Constituent	Fluid Sludge (Orchard Grass) (1973-76)		Air-Dried Sludge (Wheat) (1974-76)	
	Loamy Sand	Silt Loam	Sand	Clay
N	3.41	1.67	4.22	4.26
P	40.40	7.60	1.41	1.14
Zn	1.60	1.42	1.19	0.78
Cu	1.90	2.32	1.00	0.65
Ni	4.52	4.38	1.18	0.65
Cr	2.86	1.81	2.53	0.95
Pb	2.50	2.63	0.68	0.31
Cd	2.14	1.57	1.93	1.07

[1] Data from Table 31.
[2] NPK average of annual means.

More than four times as much nickel leached out of fluid sludge amended soils (highest loading rate) as from the NPK controls on both loamy sand and silt loam. Chromium, lead and cadmium leached from fluid sludged soils at approximately double the rate of that from NPK controls. The amounts of metals leached from air-dried sludges applied to clay soils was generally less than from NPK control. On sand soil only chromium leached out of sludge amended soil at more than double the NPK rate with cadium leaching at nearly double the NPK rate.

Table 33 compares the maximum amounts of nutrients and heavy metals recovered in leachate with the amounts applied at the highest sludge loading rate. Whereas only 2 to 4% of nitrogen applied as fluid sludge is recovered in leachate, approximately 33% of the nitrogen applied as air-dried sludge ends up in the drainage water. Of all the heavy metals monitored, only nickel and cadmium recoveries exceed 1%, with nickel recovery averaging 6.2% from fluid sludges, and 1.8% from air-dried sludges.

TABLE 33. RECOVERY EFFICIENCY (%) OF NUTRIENTS AND HEAVY METALS IN LEACHATE FROM SLUDGE AMENDED SOILS AT MAXIMUM LOADING RATE

Constituent	Fluid Sludge (Orchard Grass) (1973-76)		Air-Dried Sludge (Wheat) (1974-76)	
	Loamy Sand	Silt Loam	Sand	Clay
N	4.12	2.01	32.4	32.7
P	0.70	0.13	0.03	0.02
Zn	0.13	0.12	0.18	0.11
Cu	0.30	0.37	0.37	0.24
Ni	6.33	6.13	2.35	1.29
Cr	0.13	0.08	0.48	0.18
Pb	0.16	0.16	0.18	0.08
Cd	1.43	1.05	1.26	0.65

5.5 Material Balances

Complete material balances for the various constituents of concern must consider all possible sources of input, and loss from the system, as well as account for the changes in inventory (storage) occurring in the sludge amended soils. Analysis of soil inventory changes was not possible during the initial years of these studies without damaging crop root systems. Soil analyses for both total and soluble constituents will be conducted during 1977 by sacrificing certain replicate lysimeters. It will then be possible to compare theoretical and actual accumulations of various elements. Theoretical material balances were calculated by subtracting measured losses from the system via leachate or plant uptake from sludge loadings. Input of nutrients and metals from precipitation (rain and snowfall) was calculated and found to be insignificant compared with quantities applied as sewage sludge. Figures 39 to 43 show material balances at the end of the 1975 growing season for certain selected constituents. It is obvious that

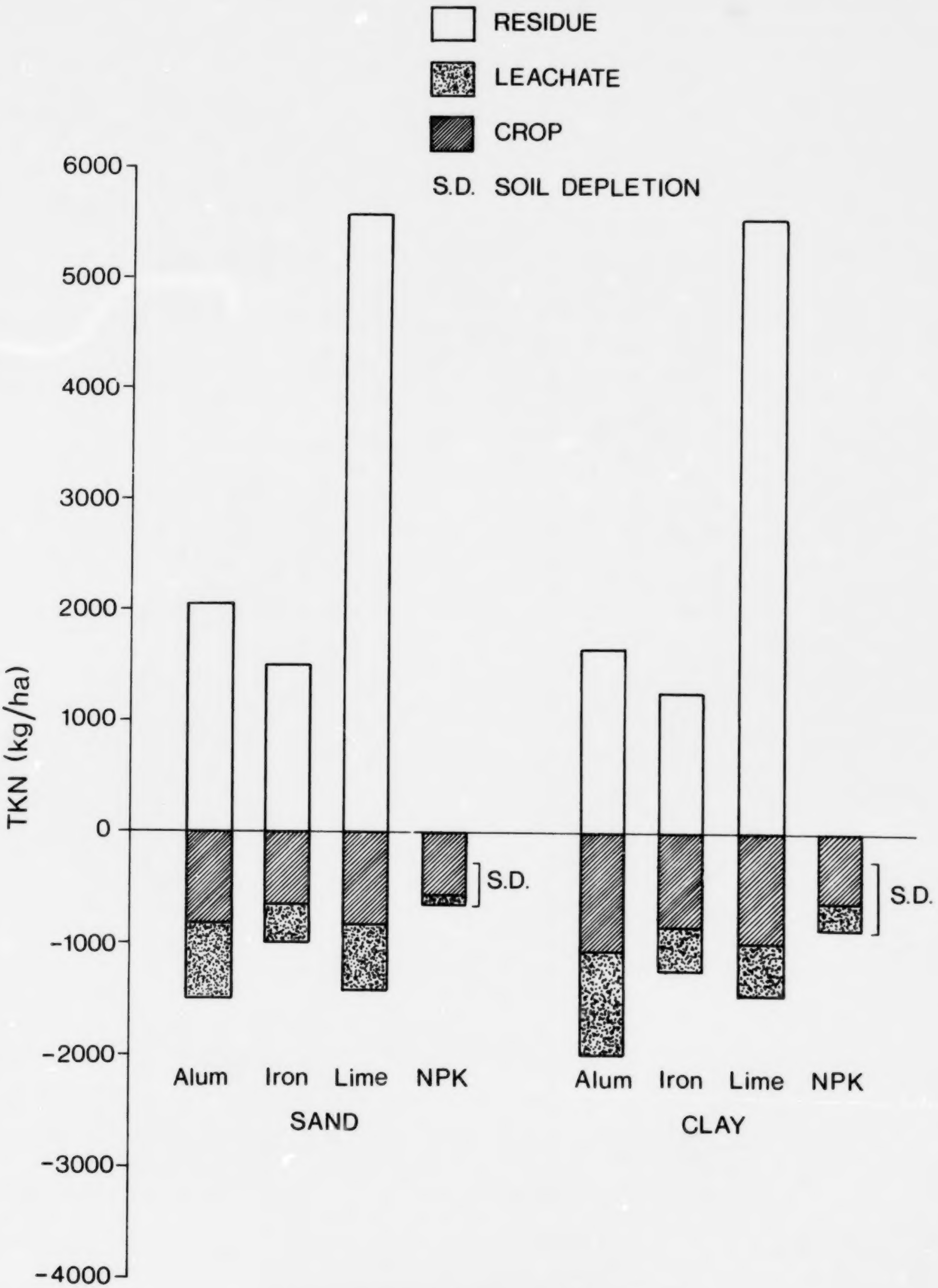


Figure 39 MASS BALANCE OF TKN (high rate)
AIR-DRIED SLUDGE-WHEAT/BUCKWHEAT

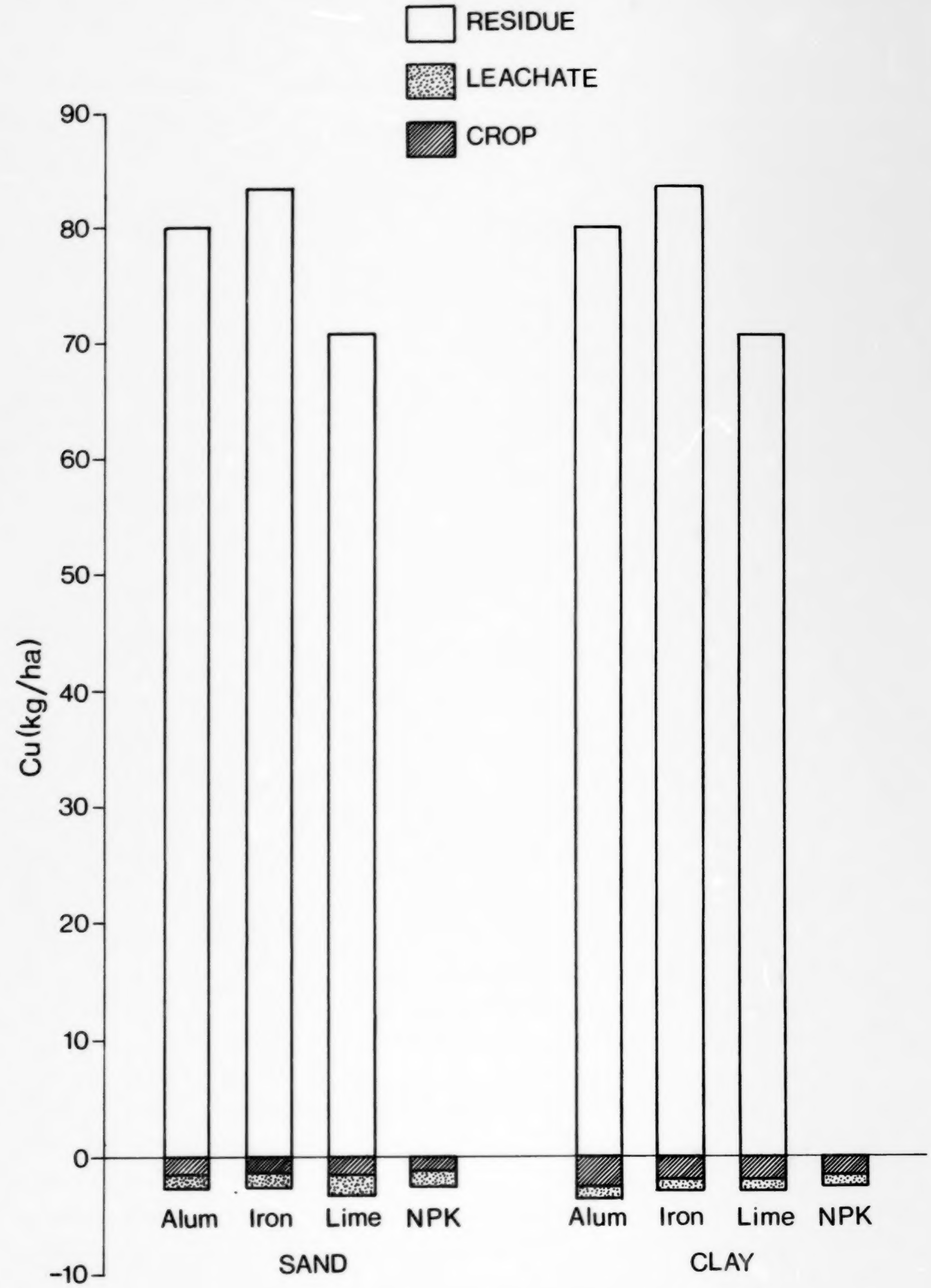


Figure 40 MASS BALANCE OF Cu (high rate)
AIR-DRIED SLUDGE -WHEAT

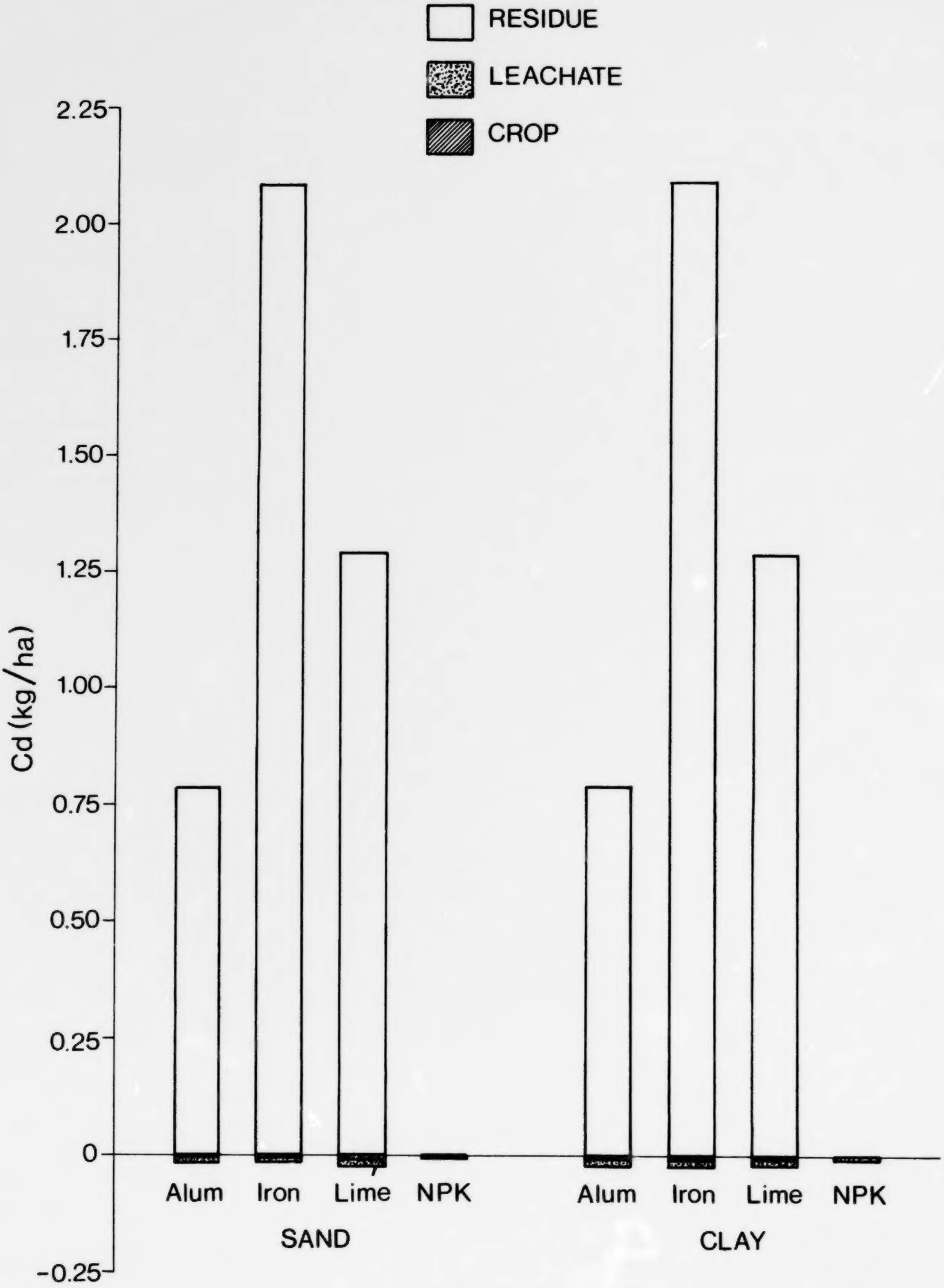


Figure 41 MASS BALANCE OF Cd (high rate)
AIR-DRIED SLUDGE-WHEAT

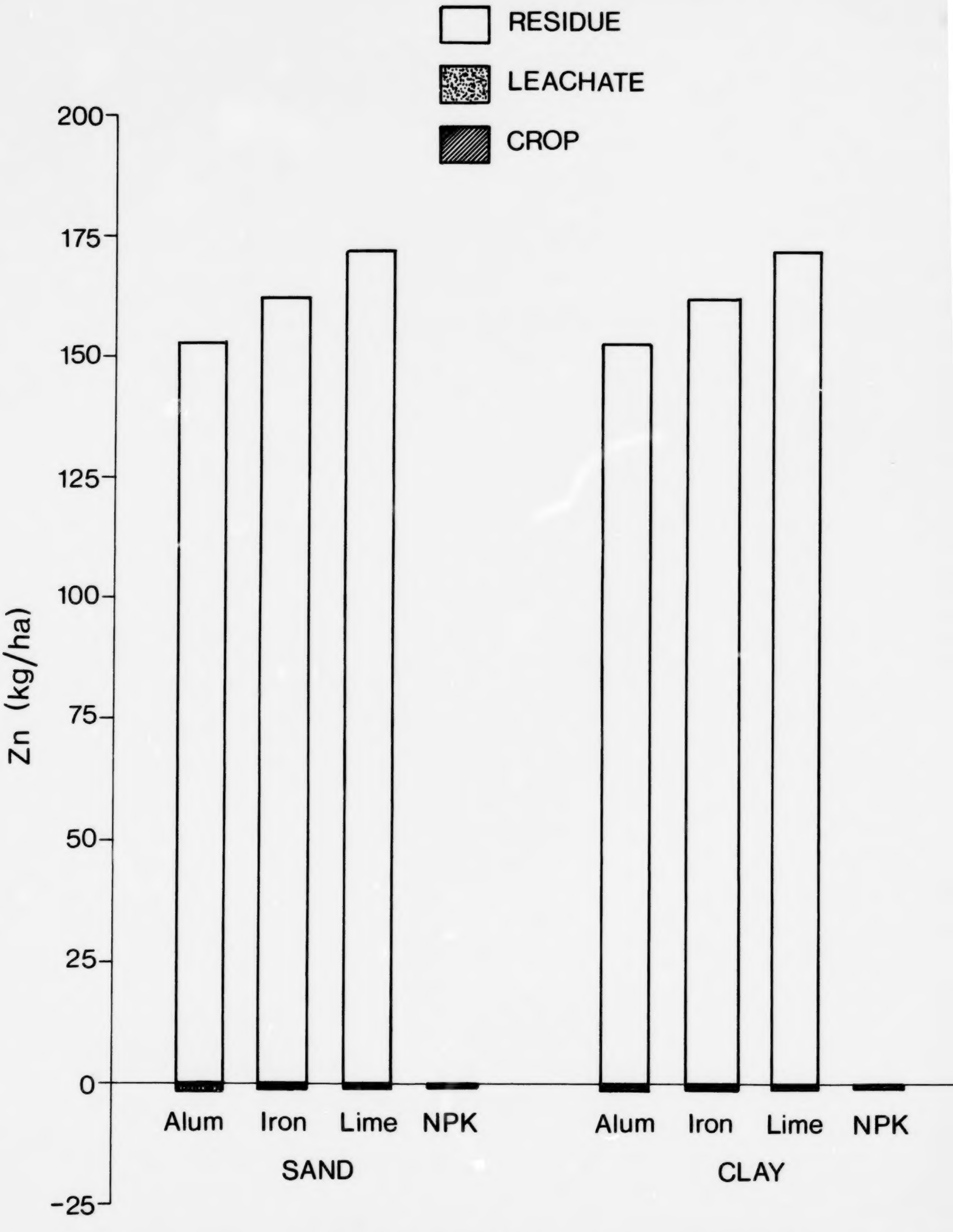


Figure 42 MASS BALANCE OF Zn (high rate)
AIR-DRIED SLUDGE-WHEAT

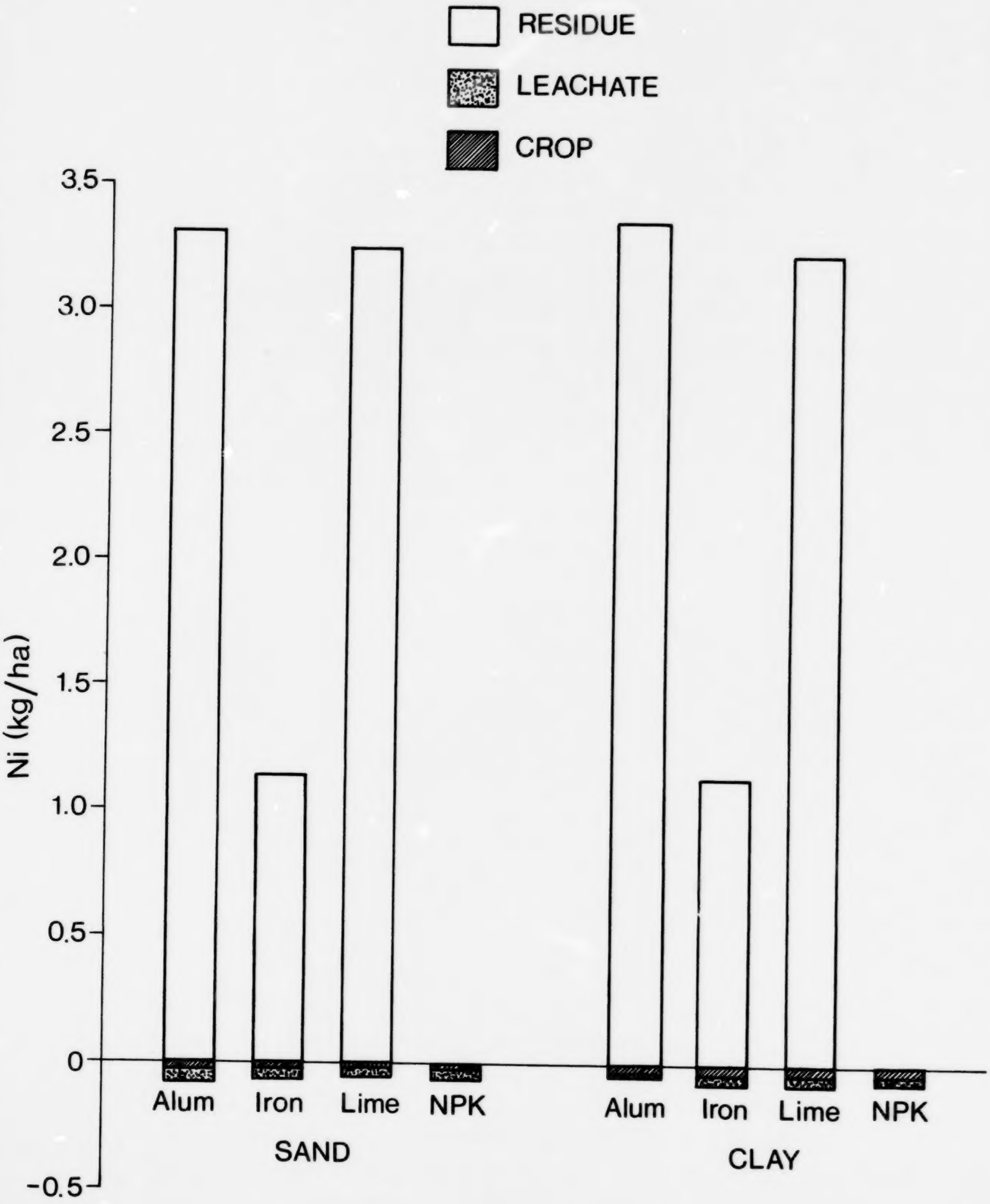


Figure 43 MASS BALANCE OF Ni (high rate) AIR-DRIED SLUDGE-WHEAT

with the exception of potassium, which is being depleted from the soil solution faster than it is being applied, the soil is the major sink for all elements being monitored. Whether or not these elements remain in the plough zone layer indefinitely or migrate vertically below the root zone as suggested by some recent studies (Kirkham, 1976), may be determined during 1977 when sludged and control soil profile samples will be analyzed by scanning electron microscopy.

5.6 Comparison of Lysimeter and Field Data

The University of Guelph has, since 1973, conducted field studies to determine the impact of fluid chemical sludges at various nitrogen loading rates on yields and plant uptake of various chemical constituents from brome grass and corn. As no provision was made for field sampling and analysis of leachate, material balances could not be calculated, but soils were periodically core sampled for nutrient and metals analyses.

Many investigators (e.g., Baxter, 1976) have commented on the difficulty of transferring data obtained from very small scale "pot" experiments in the greenhouse to actual field conditions. No data comparing the transferability and scale-up applicability of pilot scale lysimeter studies to full scale field conditions are available.

The forage crops used in Guelph (brome grass) and Burlington (orchard grass) were similar enough to allow for comparison of plant yield and uptake data. The parallel time frame, soils, sludge sources and loading rates, make a comparison of the WTC lysimeter studies and University of Guelph field studies of general interest. The quantities of nutrients and heavy metals applied to the University of Guelph field site over three years (1973 to 1975) are compared to the loadings of these same constituents to WTC lysimeter soils (Table 34). Nitrogen at the highest loading rate was 33% higher in the field study than the lysimeter N loadings. Loadings of the various constituents were reasonably similar for both iron and lime treated sludges. Most constituent loadings from alum treated sludges were similar in both field and lysimeter experiments with the exception of Ni and Cr loaded 11.3 and 35.0 times greater, respectively, in the field experiment.

TABLE 34. RATIO OF FLUID SLUDGE LOADING RATES (UNIVERSITY OF GULEPH-FIELD/ WTC-LYSIMETER) TO SILT LOAM SOIL (1973 to 1975) AT THE HIGHEST LOADING RATE

Constituent	Sludge Loading Ratios (kg/ha) University of Guelph-Field / WTC-Lysimeter		
	Alum	Iron	Lime
TKN	1.33	1.33	1.33
NH_4-N	1.90	1.50	1.49
Total P	1.14	2.53	1.69
Zn	4.35	1.50	1.39
Cu	1.27	1.43	1.29
Ni	11.30	1.90	1.97
Cr	35.0	0.98	1.14
Pb	1.64	1.35	1.03
Cd	1.20	1.03	0.80
Total Solids (tons/ha)	1.26	1.27	1.50

5.6.1 Crop yields

Response of orchard grass and brome grass to N loadings were similar in both experiments. The cumulative residual yield effects of sludge loadings (compared to NPK control) increased in the third and fourth years of both experiments. Absolute yields at the highest loading rate were approximately three times greater from lysimeter experiments. For example, the cumulative yield (high rate iron sludge) of orchard grass at the end of the third year exceeded 60 tons dry matter/ha compared to approximately 20 tons/ha for field trial harvest of brome grass. These absolute differences in crop yield are attributed to a combination of ample irrigation, geographic location (degree-days) and less competition for light from the sides of the lysimeters compared to the field plots. In view of this yield difference, actual uptake (kg/ha) of specific metals would be significantly less in the field than from lysimeters if metals concentrations were approximately equal in both orchard and brome grass samples.

5.6.2 Crop heavy metals concentration

Table 35 compares the average annual concentrations of Cd, Cr, Cu, Ni, Pb and Zn in plant tissue (mean of high rate alum iron and lime sludge treatments) from:

- WTC lysimeter study - orchard grass (1973 to 1976).
- University of Guelph field study - brome grass (1973 to 1975).
- University of Guelph greenhouse study - rye grass (1st to 6th crops).

The greenhouse "pot" studies were loaded at much higher rates of heavy metals than either the lysimeter or field studies. Salts did build up initially in the pots and lack of drainage may have affected soil oxidation reduction potential. Heavy leaching of excess salts was therefore instituted after every second crop. Despite the generally higher metal concentration values observed in the rye grass compared with field and lysimeter results, the time trends were of sufficient interest to include in this comparison.

Cadmium. Lysimeter values declined with time to 0.10 μg/g in 1976. Field studies results at the end of the third year were virtually identical with first year results (0.24 μg/g). Greenhouse results declined between the first and sixth crop to 0.40 μg/g. Deviation of Cd from NPK control values never exceeded +0.10 μg/g in either lysimeter or field tests. Although absolute values of Cd tended to reflect the relative loading of this metal in each experiment, cumulative sludge applications did not reveal time trends which approached the maximum suggested tolerance level (3.0 μg/g).

Chromium. Chromium trend results were inconsistent, fluctuating between 0.6 and 2.0 μg/g in the lysimeter experiment, compared to a range of 0.8 to 2.4 μg/g in the field experiments. Greenhouse values were not reported for the third and fifth cut as there were no significant differences between treatments. Deviations from NPK control values were generally low, rarely exceeding +0.30 μg/g. In view of the previous discussion concerning toxicity of Cr to plants and animals fed

TABLE 35. COMPARISON OF HEAVY METALS CONCENTRATIONS IN PLANT TISSUES FROM WTC LYSIMETER AND UNIVERSITY OF GUELPH STUDIES - MEAN OF ALUM, IRON AND LIME SLUDGE TREATMENTS AT THE HIGHEST RATE (μg/g dry matter)

Constituent	WTC - Lysimeter (Orchard Grass)				University of Guelph Field (Brome Grass)			University of Guelph Greenhouse[2] (Rye Grass)			
	1973	1974	1975	1976[1]	1973	1974	1975	1st Crop	3rd Crop	5th Crop	6th Crop
Cd	0.55	0.28	0.10	0.10	0.23	0.15	0.24	0.74	0.52	0.55	0.40
Cr	0.6	1.4	0.6	2.0	1.2	2.4	0.8	1.9	NA	NA	1.0
Cu	7.9	9.2	7.9	9.4	13.4	10.9	16.0	17.5	12.5	18.0	24.0
Ni	1.3	0.9	1.6	1.1	1.0	1.4	1.9	2.9	1.3	2.5	2.6
Pb	8.1	9.4	6.3	11.3	2.2	NA	2.2	4.1	5.7	2.3	1.5
Zn	48.0	65.3	58.7	58.0	31.0	30.0	46.0	54.0	66.7	58.0	94.0

[1] 1976 - 2 of 5 cuts analysed.

[2] Greenhouse study: alum sludge = Kitchener; lime sludge = Midland; iron sludge = North Toronto.

NA = not available.

crops grown on sludge amended soils, as well as recent evidence (Underwood, 1971) of a potential dietary deficiency of Cr in humans, the absolute values and trends observed in all three experiments cannot be considered a cause for concern at this time.

Copper. Copper concentration showed a slight tendency to increase with time. The highest average annual values for all three sludges analyzed in both lysimeter and field studies were below Page's (1974) suggested toxic level of 20 µg/g. The 6th rye grass crop value, however, exceeded the suggested toxic level. Deviations from NPK control values never exceeded 5 µg/g in the lysimeter study, and declined in the 1975 field study to <2 µg/g. High concentrations of copper in rye grass grown on a soil receiving unusually high metal concentrations (Fergus, Ontario) were implicated as the cause of crop yield depression (Bates et al, 1977). Although no yield depression effects have been observed to date at the highest loading rate to either lysimeter or field crops, cumulative sludge application up to the loading applied with the Fergus sludge are required to determine whether copper toxicity can be induced in forage crops by applying "reasonable" amounts of fluid sludge year after year to the same site.

Nickel. Nickel concentration ranged from 0.9 to 1.6 µg/g in the lysimeter experiment and 1.0 to 1.9 µg/g in the field experiment. Although nickel concentration appeared to increase with time in the field results, recent information from the University of Guelph (Soon, 1977) indicates that the nickel concentration of the 1976 brome grass crop (third cut) declined to less than 1.0 µg/g. Although the maximum suggested tolerance level for nickel is 3.0 µg/g (Melsted, 1973), concentrations as high as 47.0 µg/g in rye grass from the University of Guelph greenhouse experiments showed no yield depression effect (Soon, 1977). In view of this information, nickel toxicity is not considered to be a problem for any of the three crops investigated in these experiments.

Lead. Lead is the only metal whose concentration is greater in orchard grass from lysimeter experiments than either field or greenhouse plant tissue concentrations. The deviations from NPK control values are less than 2 µg/g in the lysimeter studies and less than 1 µg/g in both field and greenhouse studies. In view of previous comments concerning adsorption of automobile exhaust lead by plants growing near major highways (McLean, 1969), and the reported low uptake of lead by most agricultural crops, this element is not considered a cause for concern for the three crops grown in these experiments.

Zinc. Zinc plant tissue concentrations increased slightly with time in the lysimeter and field studies and showed a more pronounced increase in the rye grass greenhouse experiment. High uptake of zinc in rye grass grown in the greenhouse has been observed (Soon, 1977) for a highly contaminated sludge from Sarnia, Ontario. Although the maximum concentration in this crop (320 µg/g) exceeded Page's (1974) suggested toxic level of 200 µg/g, no yield depression was observed. As zinc has been implicated in the literature as being highly toxic to many plants and the University of Guelph greenhouse rye grass experiment indicates what may occur with additional sludge application to both lysimeters and field experiments, this metal will continue to be monitored in orchard grass and brome grass for at least a decade.

5.6.3 Uptake of contaminants from selected sludge farms

The applicability of research data concerning uptake of metals and other contaminants by plants grown on sludge amended soils increases in direct proportion to the experimental design size. The length of time required to determine trends which may be cause for concern is considerable. A field monitoring study was, therefore, initiated in 1976 at the Wastewater Technology Centre under the auspices of the Canada-Ontario Agreement on Great Lakes Water Quality to determine whether continuous sludge application to soils at selected sludge farms in Ontario for at least five years increased the uptake of As, Cd, Co, Cr, Cu, Hg, Mo, Ni, Pb, Zn or PCB's by wheat, corn, or oats compared to adjacent control fields that never received sludge.

While data reduction and analysis has not yet been completed, initial information (Monteith, 1977) indicates the following:

Arsenic (As)

Concentration in soils 1.0 to 7.2 μg/g (dry weight).

Concentration in plants 0.1 to 0.8 μg/g (dry weight).

No increased uptake of As due to sludge application was evident at any of the ten farms monitored.

Cadmium (Cd)

Concentration in soils 0.3 to 1.2 μg/g.

Concentration in plants 0.02 to 0.74 μg/g.

Only one of ten crops sampled (corn stover) showed significant uptake on "sludged" versus "control" site. Maximum plant concentration observed (0.74 μg/g), was well below the suggested tolerance level of 3.0 μg/g.

Cobalt (Co)

Concentration in soils 24 to 64 μg/g.

Concentration in plants 1.8 to 3.3 μg/g.

No significant increase in Co uptake from any sludge application compared to "control" crop could be discerned.

Chromium (Cr)

Concentration in soils 40 to 235 μg/g.

Concentration in plants 1.4 to 18.0 μg/g.

Only one crop (corn stover) showed significant increase in Cr uptake from "sludged" versus "control" fields. No yield depression at the highest Cr plant tissue concentration was observed.

Copper (Cu)

Concentration in soils 15 to 69 μg/g.

Concentration in plants 1.6 to 14.0 μg/g.

Seven of ten crops showed evidence of increased copper uptake from "sludged" versus "control" fields. No yield depression at highest Cu concentration was observed.

Mercury (Hg)

Concentration in soils 0.03 to 0.20 μg/g.

Concentration in plants 0.01 to 0.026 μg/g.

Only two crop samples (corn stover) showed slight increase in Hg from "sludged" versus "control" site. No yield depression at highest Hg concentration was observed.

Molybdenum (Mo)

Concentration in soil 2.5 to 21.0 μg/g.

Concentration in plant 0.4 to 23.8 μg/g.

Only one sample (corn stover - Stratford, Ontario) showed evidence of molybdenum uptake above the suggested maximum tolerance level of 3.0 μg/g (Melsted, 1973). High Mo in corn was not phytotoxic. Possible Molybdenosis of cattle fed high Mo forage crops is, however, of concern.

Nickel (Ni)

Concentration in soil 10 to 45 μg/kg.

Concentration in plants 1 to 11 μg/kg.

No significant increase in Ni uptake from any sludge application compared to "contol" crop could be discerned.

Lead (Pb)

Concentration in soil 13 to 97 μg/kg.

Concentration in plants 0.3 to 1.1 μg/kg.

No significant increase in Pb uptake from any sludge application compared to control crop could be discerned.

Zinc (Zn)

Concentration in soil 95 to 415 μg/kg.

Concentration in plants 7.5 to 113 μg/kg.

Only one crop (wheat straw) showed any significant increase in Zn uptake from sludge application compared to "control" crop. No evidence of yield depression at highest plant concentration was observed.

PCB's

Concentration in soil 1 to 125 ppb (μg/kg).

Concentration in plants 7 to 83 ppb (μg/kg).

No criteria exist for maximum suggested tolerance level in plants. The Canadian temporary guideline for PCB's in commercial fish is several orders of magnitude greater than maximum concentration observed in plant samples in this study (NH & W, 1975). No significant increase in PCB's from sludge application compared to control could be discerned.

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ABBREVIATIONS

a = acre
Al = aluminum
As = arsenic
Ave = average (arithmetic mean)
BOD = 5-day biochemical oxygen demand
°C = degrees celsius
Ca = calcium
Cd = cadmium
Cl = chloride
cm = centimeter
COA = Canada-Ontario Agreement
Co = cobalt
Cr = chromium
Cu = copper
CCE = carbon chloroform extract (organic matter)
CEC = cation exchange capacity (milliequivalents/100 grams)
Fe = iron
g = gram
GCSD - Greater Chicago Sanitary District
ha = hectare
Hg = mercury
H_2O = water
K = potassium
kg = kilograms
m^2 = square metre
m^3 = cubic metre
mg = milligram (10^{-3} gram)
ml = millilitre (10^{-3} litre)
Mn = manganese
Mo = molybdenum
MAC = maximum allowable concentration
MPC = maximum permissible concentration
MTL = maximum tolerance level

N = nitrogen

Na = sodium

NH_4-N = ammonium nitrogen

Ni = nickel

NO_2-N = nitrite nitrogen

NO_3-N = nitrate nitrogen

NPK = commercial fertilizer (nitrogen, phosphorus, potassium)

OMAF = Ontario Ministry of Agriculture and Food

OMOE = Ontario Ministry of the Environment

P = phosphorus

Pb = lead

PCB's = polychlorinated biphenyls

Se = selenium

TKN = total Kjeldahl nitrogen

TOC = total organic carbon

ton = metric ton (1000 kg)

μg = microgram (10^{-6} gram)

USDA = United States Department of Agriculture

USEPA = United States Environmental Protection Agency

WHO = World Health Organization

WPCP = water pollution control plant

WTC = Wastewater Technology Centre (Burlington)

$\bar{x}$ = arithmetic mean

yr = year

ZE = zinc equivalent

Zn = zinc